

# The American Mineralogist

## *Journal of the Mineralogical Society of America*

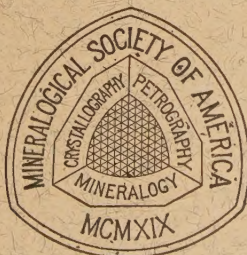
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Nos. 3 and 4

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# THE AMERICAN MINERALOGIST

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## SYNTAXIC INTERGROWTHS IN THE ANDORITE SERIES\*

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### ABSTRACT

Most specimens of andorite are syntaxial intergrowths of two distinct species, which have also been observed separately. Rather than revive the name sundtite, we propose to call them andorite IV and andorite VI because their  $c$  translations are multiples,  $4c'$  and  $6c'$ , of a common pseudo-period  $c' = 4.29 \text{ \AA}$ . They have identical  $a$  and  $b$  translations, equal to 13.03 and 19.15  $\text{\AA}$  respectively. They have different morphological developments, already noted by Ungemach, with morphological aspects  $P^*ca$  and  $Pn^*a$  referred to the true cells. Weissenberg and precession  $x$ -ray patterns yield diffraction aspects  $P^{**}a$  and  $Pn^*a$ ; they also show a strikingly marked pseudo-aspect  $Bb^{**}$ , for the pseudo-cell  $abc'$ , common to both species. Specific gravity determinations on polycrystals give values ranging from 5.23 to 5.44, suggesting different chemical compositions for the two species. Ramdohrite = andorite VI.

### INTRODUCTION

Andorite  $\text{PbAgSb}_3\text{S}_6$  was first found at Felsöbánya and was described by Krenner (1892). Sundtite, announced as  $(\text{Ag}_2, \text{Cu}_2, \text{Fe}) \text{Sb}_2\text{S}_6$  by Brögger (1893), came from Oruro, Bolivia. Stelzner (1895) described a mineral under the name webnerite. Prior and Spencer (1897) re-examined these minerals and concluded that both sundtite and webnerite were identical with Krenner's andorite, with up to 2 per cent copper substitution in the case of sundtite. Ahlfeld (1930) described a mineral of composition  $\text{Pb}_3\text{Ag}_2\text{Sb}_6\text{S}_{13}$  or  $\text{Pb}_6\text{Ag}_4\text{Sb}_{10}\text{S}_{23}$  from Potosí, Bolivia, and named it ramdohrite. It is known to have a powder pattern "very similar" to that of andorite (Strunz, 1949).

Ungemach studied the Oruro material, on which he noted two distinct types of morphological development. On this basis he suggested that

\* Publication authorized by the Director, U.S. Geological Survey.

† This paper presents some of the unpublished data discussed in the presidential address *Edifices of Periodic Matter*, which was delivered in Toronto, on November 10, 1953, to the Mineralogical Society of America.



sundtite should be reinstated. His results are mentioned in the literature (Ungemach, 1923) though not fully discussed. Preliminary *x*-ray results obtained by the senior author (J. D. H. Donnay, 1941) showed that the andorite-sundtite problem was still unsolved and needed further study. Nuffield (1945) gave cell dimensions and space groups for andorite, ramdohrite, and fizelyite.

#### MORPHOLOGICAL RESULTS

Ungemach's notebooks, which had been bequeathed by him to one of us (JDHD), were carefully examined. He had measured 24 "crystals" from Oruro. Although he was not aware that nearly all of them are syntaxic intergrowths of two species (see below, under *x*-ray results), he separated his specimens into two types, which he considered distinct species. His conclusion rested on two facts, which an admirable power of observation and patient scrutiny had enabled him to establish, namely: (1) differences in the morphological developments of the two types of "crystals"; (2) small angular differences in their interfacial angles.

For the type of crystals described by Brögger, Ungemach found the axial ratios:

$$0.6773:1:0.4429.$$

He referred to them as "sundtite" (=our andorite IV, see below). For other crystals, which he called "andorite" (=our andorite VI), he multiplied the *c/b* ratio by 3 and gave the numerical values:

$$0.6807:1:1.3346 = 0.6807:1:0.4449 \times 3.$$

The axial ratios given for andorite in the literature (Strunz, 1949),

$$0.6771:1:0.4458,$$

are those of Brögger (1893).

The only emendations required by Ungemach's conclusions stem from the fact that the edifices he measured, although they simulated crystals of a single species, actually were intergrowths of crystals of two species in parallel orientations. The term *polycrystal* (G. Donnay, 1953) can be applied to such an intergrowth.

The numerical values reported by Ungemach for the axial ratios will probably have to be refined because some of the interfacial angles used in the calculations were due to the species intergrown with the species under consideration. For example on a polycrystal in which andorite IV was predominant, and which Ungemach accordingly treated as an andorite IV crystal, he would have included the angle (001):(091) among those on which to base his *c/b* ratio, and yet (091) would have been a face of andorite VI.

Ungemach stated that Brögger's (331) must be chosen as the new unit



face in the case of andorite VI. We confirm this statement by morphological analysis. All the crystals of andorite VI plainly show the morphological aspect  $Pn^*a$ , with Brögger's  $c$  trebled (Fig. 1), so that the morphological space group is either  $Pnma$  ( $D_{2h}^{16}$ ) or  $Pn2a$  ( $C_{2v}^9$ ).

Ungemach also stated that Brögger's unit face (111) was correctly chosen for andorite IV. Our own morphological analysis, on the contrary,

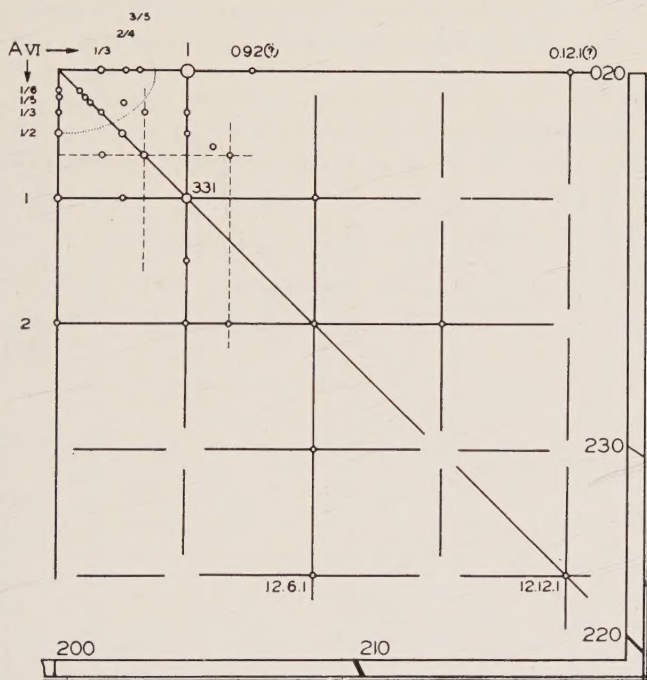


FIG. 1. Gnomonic projection<sup>‡</sup> of a polycrystal, dominantly composed of andorite VI, referred to Brögger's axial elements. The pole indexed 331 in Brögger's notation should be taken as 111 for andorite VI. Some poles, connected by dashed lines, belong to andorite IV. The space group can be read directly from the projection ( $Pnma = D_{2h}^{16}$ ). Ungemach's specimen B425.

indicates that the face to be selected as the new unit face is Brögger's (221). The first layer of the reciprocal lattice of andorite IV is then remarkably brought out, on the gnomonic projections of all the polycrystals on which andorite IV predominates, by such faces as Brögger's (221), (241), (261), (281), (441), (481), which can be seen on Fig. 2, and (2.12.1), (661), (12.12.1), (24.12.1), (12.6.1), which were observed on other speci-

<sup>‡</sup> The gnomonic projections (Figs. 1-3) have been distorted by shrinking the  $a^*$  direction in the ratio of 3 to 2, so that the rectangular mesh can be represented by a square. The sphere of projection becomes an ellipsoid.



mens. On some crystals (Fig. 2) it may be difficult to decide whether the aspect is  $P^{*}a$  or  $P^{*}ca$ . On the whole, however, the frequency data collected on the  $(h0l)$  forms definitely point to Brögger's (101) = new (102) as the dominant face in the zone. The zone of the  $(h0l)$  faces, moreover, is of the simple type, as the next frequent faces are (in the new notation) (104) and (206), and the multiple indices of the latter are confirmed by (308) and (3.0.10), both observed. The zone of the  $(hk0)$  faces is an outstanding example of a simple zone with (210) dominant (Fig. 2). The

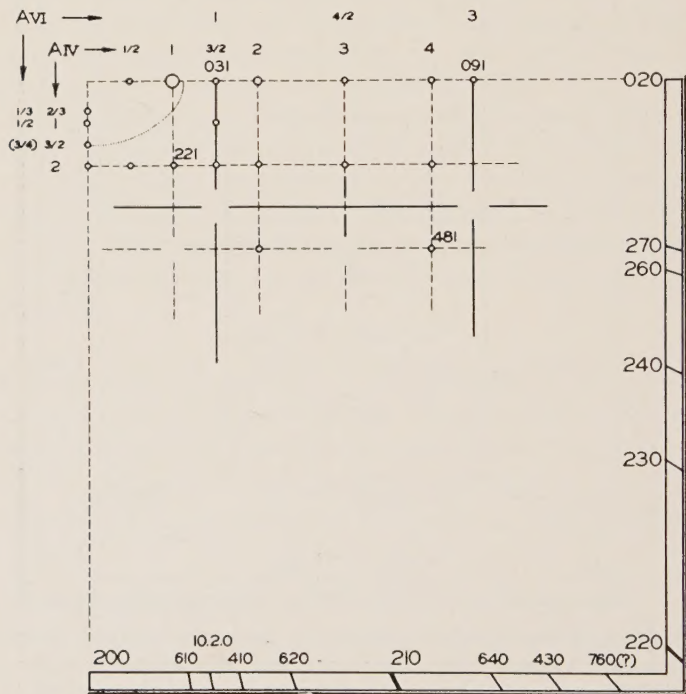


FIG. 2. Gnomonic projection of nearly pure andorite IV, referred to Brögger's axial elements. The pole indexed 221 in Brögger's notation becomes 111 for andorite IV. Pole 091 belongs to a double series (031-061-091) due to andorite VI. The zone of the  $h0l$  poles can be interpreted either as simple with unit face dominant (mirror) or as simple with dominant face shifted toward the  $c$  face ( $c$  glide plane). The morphological space group is read as  $Pmma(D_{2h}^5)$  or  $Pmca(D_{2h}^{11})$ . Ungemach's specimen A356.

morphology thus unmistakably indicates aspect  $P^{*}ca$ , and the morphological space group is either  $Pmca(D_{2h}^{11})$  or  $P2ca(C_{2v}^5)$ .

The gnomonic projection plotted for the original andorite, according to the data published by Krenner (1892), shows that both andorite IV and andorite VI are present in an intergrowth but that andorite IV predominates. Using Brögger's notation we see that  $q(221)$ ,  $r(121)$ , and  $\gamma(021)$



belong to andorite IV, whereas  $\beta(131)$  and especially  $\gamma(031)$  and  $t(091)$  can be referred only to andorite VI. Some faces, such as  $\alpha(011)$  and all the  $(hk0)$  faces, are compatible with either species. Koch (1928) studied new material from Felsöbánya. His data confirm the above conclusion, based on Krenner's results. He found some new forms, among which  $\pi(032)$ , a rather frequent one, will find its place as a secondary form of andorite VI to be expected between  $\gamma$  and  $\alpha$ .

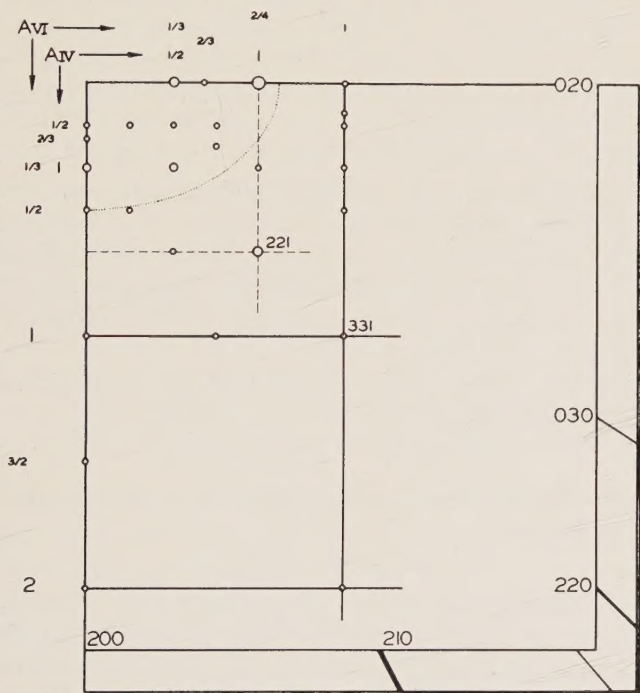


FIG. 3. Gnomonic projection of a polycrystal of andorite VI (zone lines solid) and andorite IV (zone lines dashed), referred to Brögger's axial elements. The zone of the  $0kl$  poles is a simple zone for andorite IV, and a double zone for andorite VI. The zone of the  $h0l$  poles shows the simple zone, with Brögger's 301 dominant, of andorite VI and another simple zone, with Brögger's 101 dominant, due to andorite IV. The zone of the  $hk0$  poles is the same for both species; it is a simple zone with 210 dominant. Ungemach's specimen B485.

Brögger's data (1893) on sundtite from Oruro yield a gnomonic projection remarkably similar to that of the Krenner specimens. Additional faces of andorite IV are  $f(101)$  and  $h(102)$ . Andorite VI acquires  $d(601)$  and  $e(302)$ , although (301) is not reported. Andorite IV is again preponderant.

From the confrontation of such similar data, the conclusion reached by



Prior and Spencer (1897), that andorite and sundtite were one and the same species, was inevitable. We were fortunate in finding, in Ungemach's material, a specimen (No. B 426) that is almost exclusively andorite VI. Its gnomonic projection displays even more forms of andorite VI than does that shown in Fig. 1. For instance—again in Brögger's notation—(091) and (061), the former larger than the latter, fit in the double zone of the  $(0kl)$  faces, while (901) and (902) complete the simple zone of the  $(h0l)$  faces. The difference between this projection and that of Fig. 2, which represents an almost pure andorite IV (No. A 356), is indeed striking. There is no doubt that, if Prior and Spencer had had such crystals available, sundtite would never have been discredited.

### X-RAY RESULTS

Rotation, Weissenberg, and precession photographs were taken, with copper and molybdenum radiations, on fragments of the polycrystals that had been measured by Ungemach, on the material studied by Nuffield (1945), and on fragments of a ramdohrite specimen, from the mine Chocaya la Vieja, Potosí, Bolivia, borrowed from the Harvard University Museum.

The lengths of  $a$  and  $b$  are the same for both species within the limits of error of our measurements,

$$a = 13.03 \pm 0.04, \quad b = 19.15 \pm 0.06 \text{ \AA}.$$

The  $c$ -axis rotation photographs show an intense layer line, which yields a pseudo-translation

$$c' = 4.29 \pm 0.02 \text{ \AA},$$

the same for both species. The small angular differences observed by Ungemach between corresponding interfacial angles of the two species, which led him to slightly different axial ratios (see above), cannot be confirmed by the present  $x$ -ray work. The first strong layer line on the  $c$ -axis rotation patterns is the fourth layer line in the case of one species, which is called andorite IV for this reason, and the sixth layer line in the case of the other species, which is accordingly designated andorite VI. The true  $c$  is therefore equal to  $4c' = 17.16 \text{ \AA}$  for andorite IV and  $6c' = 25.74 \text{ \AA}$  for andorite VI. Although the intermediate layer lines are clearly visible on a 2-hour rotation photograph superimposed on a 2-hour  $10^\circ$ -oscillation photograph,<sup>1</sup> exposures of the order of 20 hours are required to bring out the weak spots on precession pictures taken with

<sup>1</sup> Such a combined photograph gives the same information as a rotation photograph if the crystal is properly adjusted. If the crystal is maladjusted, the arc corrections can be obtained from the film so that it is not necessary to take a separate adjustment picture (Donnay and Donnay, 1952).



nickel-filtered copper radiation. The Weissenberg photographs of the strong layer lines were exposed for 22 hours; those of the weak layer lines for twice that time.

The reflections that lead to the pseudo-cell  $abc'$  show "systematic" omissions, so that the space group of the pseudo-cell can be determined. Let  $l'$  designate the index that refers to the pseudo-translation  $c'$ . The following reflections are observed:  $hkl'$  with  $(h+l')$  even;  $0kl'$  with  $k$  and  $l'$  both even. The diffraction aspect is therefore  $Bb^{**}$  and the space group of the pseudo-cell is  $Bbmm$  ( $D_{2h}^{17}$ ),  $Bb2m$  ( $C_{2v}^{12}$ ), or  $Bbm2$  ( $C_{2v}^{16}$ ). The diffraction aspect is the same for both species. Inasmuch as the omissions considered in its determination are due to features of the crystal structure, that is, are *structural extinctions* rather than *space group extinctions*, the aspect of the pseudo-cell is only a pseudo-aspect. The pseudo-symmetry is so pronounced in the present case, however, that the pseudo-aspect of the pseudo-cell could safely be assumed in all but the final stages of a crystal-structure determination. The reflections due to the pseudo-cells of the two species are so nearly identical that the crystal structures must only differ in details.

Systematic omissions among reflections indexed with respect to the true cells were obtained from zero-layer and first-layer  $c$ -axis Weissenberg patterns, for the  $hk0$  and the  $hk1$  reflections, and from precession patterns, for the  $0kl$  and the  $h0l$  reflections. The following reflections were observed.

In andorite VI:  $hkl$  present in all orders;  $0kl$  with  $(k+l)$  even;  $h0l$  in all orders;  $hk0$  with  $h$  even. The diffraction aspect is therefore  $Pn^*a$ , and the possible space groups  $Pnma$  ( $D_{2h}^{16}$ ) and  $Pn2a$  ( $C_{2v}^9$ ).

In andorite IV:  $hkl$  present in all orders;  $0kl$  in all orders;  $h0l$  in all orders;  $hk0$  with  $h$  even. The diffraction aspect is  $P^{**}a$ , compatible with three space groups:  $Pnma$  ( $D_{2h}^5$ ),  $P2ma$  ( $C_{2v}^2$ ), and  $Pm2a$  ( $C_{2v}^4$ ).

Although the observations have been given for each species separately, most films showed the superimposed patterns of the two species in syntaxic intergrowth. All the specimens used by Nuffield were polycrystals, which accounts for his erroneous interpretation (Fig. 4). What would be called "andorite XII" in our nomenclature does not exist, and the systematically missing layer lines are no longer puzzling. Dr. Nuffield has re-examined his films and confirms our findings (personal communication).

A ramdohrite crystal from the type locality, Potosí, Bolivia, was studied by Nuffield (1945), who reported cell dimensions identical, within limits of error, with those of our andorite VI. We obtained crystals from the same locality and confirmed the identity of ramdohrite and andorite VI. No andorite IV was found on the specimen. We also confirmed Nuf-

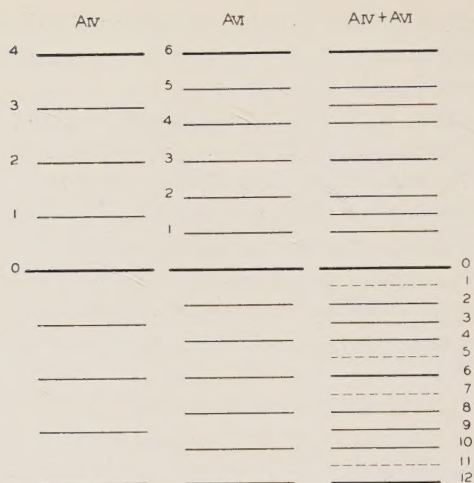


FIG. 4. Schematic representation of layer lines on rotation  $x$ -ray patterns. The pseudo period of  $4.29 \text{ \AA}$  along the  $c$  axis is shown by the first strong layer line, which is the fourth one in andorite IV and the sixth one in andorite VI. The effects observed with a polycrystal (IV+VI) are due to the superposition of the two patterns; they could be mistaken for those of a single crystal, in which the first strong line would be the twelfth one and in which the systematically absent first, fifth, seventh, and eleventh lines would be ascribed to structural extinctions.

field's results on fizelyite, which is definitely not an "andorite II" (Donnay and Donnay, 1952).

No material described as webnerite was available to us or to Nuffield.

Gnomonic projections of all the Ungemach's measured polycrystals were plotted, and the zone lines of the two species drawn with solid and dashed lines (cf. Figs. 1 to 3). In this way it was possible to see which of the two species controlled the morphological development and was presumably dominant. The  $x$ -ray photographs of the same polycrystals did not in every case confirm the morphological conclusion. The reason for this is simple: the  $x$ -ray beam may penetrate only one crystal, especially when the tip of the edifice, which is usually bathed in the beam, is homogeneous or happens to contain a preponderance of the species that is the minor constituent of the polycrystal.

The comparison of the  $x$ -ray results with the morphological ones shows that Brögger's axial elements,

$$0.6771:1:0.4458,$$

are those of a cell twice as high as the pseudo-cell, which has height  $c' = 4.29 \text{ \AA}$ . For Brögger's cell our  $x$ -ray results yield the ratios

$$0.680:1:0.448.$$



This cell probably represents the best compromise if one tries to refer the forms of both species to a single cell, but it is inadequate to express the morphology of either species. The true cell governs the morphological development in each species. The morphological space groups are the same as the structural space groups, with one deviation—the morphology of andorite IV demands a  $c$  glide plane perpendicular to the  $(h0l)$  faces, whereas its crystal structure requires either a mirror or no symmetry plane at all. It is admittedly hard to see how a structural mirror would cease to be a mirror when morphology is concerned. This anomaly suggests the absence of symmetry planes normal to the  $b$  axis in the crystal structure and thereby makes the space group  $Pm2a$  the most likely one.

#### REMARKS ON CHEMICAL COMPOSITION

The very occurrence of two species in syntactic intergrowth was taken by Ungemach (1923) to mean that they were polymorphic forms (*polytypes*), but it is now known that such is not necessarily the case. It is true for carborundum (Ramsdell, 1947; Donnay, 1943) and for zinc sulfide (Fronzel and Palache, 1950). In the bastnaesite-synchisite series, however, chemically different species have been found to intergrow (Donnay and Donnay, 1953). In that series of minerals (bastnaesite, parisite, roentgenite, synchisite), a pseudo-cell is common to all the species though one of its dimensions—namely its  $c$  length—varies from species to species. In the present case of andorite IV and andorite VI, there also exists a common pseudo-cell, but we have not been able to detect a variation in any of its dimensions. At first sight, this situation would seem to favor the assumption of identity of chemical composition. The density, of course, would have to be constant too, and it provides a way of attacking the problem of the composition. The available chemical analyses vary a great deal; that of ramdohrite (by Brendler, on type material) departs considerably from the others.

Five new determinations of the specific gravity were made on polycrystals from Oruro by means of the Berman balance. They are: 5.23 (on 31 mg), 5.23 (on 14 mg), 5.33 (on 8 mg), 5.36 (on 19 mg), 5.44 (on 20 mg). The last one was obtained on material that was predominantly andorite VI, judging from its gnomonic projection. One of the 5.23 values was obtained on 31 mg of material in which andorite IV was preponderant. The relative proportions of andorite IV and andorite VI in the other polycrystals cannot be estimated as easily. The highest value in the literature is 5.43, reported for ramdohrite by Fronzel (in Nuffield, 1945). The values found by Prior and Spencer (1897) were 5.38 (Oruro) and 5.33 (Felsöbánya). This range of specific gravities suggests that the compositions of andorite IV and andorite VI may be different.

The pseudo-cell  $abc'$  has volume  $V' = 1070 \text{ \AA}^3$ . With 4 formula units of composition  $\text{PbAgSb}_3\text{S}_6$ , the calculated specific gravity is 5.41. If it were to contain 1 formula unit of  $\text{Pb}_6\text{Ag}_4\text{Sb}_{10}\text{S}_{23}$ , one of the compositions suggested for ramdohrite, the calculated specific gravity would be 5.63, 4% higher than the maximum value observed. This means either that no pure andorite VI specimen has as yet been used for specific gravity determination or that this formula is not the correct one. The alternate formula for ramdohrite,  $\text{Pb}_3\text{Ag}_2\text{Sb}_6\text{S}_{13}$ , is even less probable, as it leads to a calculated specific gravity as high as 6.16. As to the lowest value observed namely 5.23, it will require a "molecular weight" equal to 3,370 in the pseudo-cell. This is 121 less than the formula weight of  $\text{Pb}_4\text{Ag}_4\text{Sb}_{12}\text{S}_{24}$ .

Chemical analyses of pure samples of each species are needed to solve the problem. In the meantime we propose to use the designations andorite IV and andorite VI to stress the structural relationship of the two species.

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# FLETCHER'S INDICATRIX AND THE ELECTROMAGNETIC THEORY OF LIGHT

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## ABSTRACT

The optical scalars and vectors for an inactive, crystalline dielectric are first deduced from the Maxwell equations for the electromagnetic field and it is then shown how these can, in fact, be derived from the Fletcher indicatrix. Attention is drawn to the importance of the focal lines in the geometry of the indicatrix.

In most texts on crystal optics in use amongst mineralogists and crystallographers at the present time the optical properties of crystals are derived from a consideration of the Fletcher indicatrix.<sup>1</sup> This has proved a most useful surface of reference as the primary concern of the mineralogist is with wave-normals, refractive indices and directions of vibration, all of which are readily derived from this simple figure. The enquiring student however always wishes to know how the surface itself is obtained and how it is related to the electromagnetic theory of light, and it is difficult to refer him to any text where the subject is concisely treated. Fletcher himself presents the indicatrix in a purely geometrical form and does not link it up with any specific view regarding the nature of light. In the systematic German texts,<sup>2</sup> although the connection of the surface with the results of the electromagnetic theory is pointed out, it is a little difficult for the student to disentangle the proof from the other, possibly more fundamental, aspects of optical theory. It seems worth while therefore to present a short statement showing how this reference surface is bound up with the classical theory of the electromagnetic nature of light.

The treatment adopted is, first, to derive in the usual way the optical scalars and vectors using Maxwell's equations, and then to show that these can in fact be derived from the indicatrix. The presentation is much simplified by using the abbreviated notation of Cartesian tensors<sup>3</sup> and, since the simpler theory of the propagation of light in isotropic media is adequately treated in standard works, a knowledge of it is assumed in what follows.

<sup>1</sup> Fletcher L.: *The optical indicatrix and the transmission of light in crystals*, London (1892).

<sup>2</sup> Pockels, F.: *Lehrbuch der Kristalloptik*, B. G. Teubner, Leipzig and Berlin (1906).  
Sivessy, G.: *Handbuch d. Physik*, vol. 20, J. Springer, Berlin (1928).

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<sup>3</sup> Jeffreys, Vide, H.: *Cartesian Tensors*, Cambridge University Press (1931).



The various quantities are represented by the following symbols:

space co-ordinates	$x_i, \xi_i$	directions-cosines	$l_i, m_i$
outward normal	$n_i$	volume	$\tau$
surface	$S$	total energy density	$w$
volume density of charge, e.s.u.	$\rho$	magnetic energy density	$t$
electrical energy density	$u$	magnetic intensity—m.s.u.	$H_i$
electrical intensity—e.s.u.	$E_i$	conduction current density	$i_i$
dielectric displacement	$D_i$	Poynting energy vector	$N_i$
magnetic induction—m.s.u.	$B_i$	dielectric constant	$K$
direction-cosines of $N_i$	$\beta_i$	velocity of electromagnetic radiation in vacuo	$c$
wave-length	$\lambda$	frequency	$\nu$
period	$T$	refractive index	$n$
ray index	$r$	wave velocity	$\alpha$
ray velocity	$\beta$	principal wave (and ray) velocities	$v', v'', v'''$
principal refractive indices	$\begin{cases} n'n''n''', \\ n' < n''' \end{cases}$		
time	$t$		

Differentiation with respect to time is indicated by a dot over the quantity concerned.

## 1. The Relations Between the Electromagnetic Quantities at a Point in a Varying Field are given by the Four Maxwell Equations

$$\begin{aligned} \text{curl } H_i &= \frac{4\pi}{c} i_i + \frac{\dot{D}_i}{c} & \text{div } D_i &= 4\pi\rho \\ \text{curl } E_i &= -\frac{\dot{B}_i}{c} & \text{div } B_i &= 0 \end{aligned}$$

In the classical continuum theory the specific properties with which we are concerned in the transmission of light by non-conducting crystals are the magnetic permeability and the dielectric constant. The first of these we take as unity on the assumption that the crystals are non-magnetic. It remains therefore to investigate the nature of the dielectric constant.

## 2. Relation Between $D_i$ and $E_i$

In an isotropic substance  $D_i$  coincides with  $E_i$  but in an anisotropic material this is no longer the case.

Thus  $E_1$  itself will produce a displacement with components along  $0x_1, 0x_2$  and  $0x_3$ . Similarly for  $E_2$  and  $E_3$ .

Each of the components of the displacement therefore will be the sum of the contributions from each of the components of the intensity, i.e.

e.g., 
$$D_1 = K_{11}E_1 + K_{12}E_2 + K_{13}E_3$$

where  $K_{12}E_2$  is the contribution of  $E_2$  to the displacement along  $0x_1$ , and so on.

In this case therefore the dielectric factor is a second order tensor and the displacement is the product of this tensor and  $E_i$  thus,

$$D_i = K_{ij}E_j.$$

### 3. The Energy Relations in an Anisotropic Dielectric

The magnetic and electrical energy densities are given by,

$$t = \frac{H_i^2}{8\pi} \quad u = \frac{E_i D_i}{8\pi} = \frac{K_{ij}E_i E_j}{8\pi}.$$

Since  $\text{div } [E, H]_m = H_i \cdot \text{curl } E_i - E_i \cdot \text{curl } H_i$ , we have, from the general equations of the electromagnetic field,

$$- \frac{(E_i \dot{D}_i + H_i \dot{B}_i)}{4\pi} = i_i E_i + \frac{c \text{div } [E, H]_m}{4\pi}$$

which here becomes,

$$K_{ij}E_i \dot{E}_j - \frac{1}{2} \dot{H}_i^2 = -c \text{div } [E, H]_m.$$

Integrating throughout any volume,

$$\frac{1}{4\pi} \int (K_{ij}E_i \dot{E}_j + \frac{1}{2} \dot{H}_i^2) d\tau = - \frac{c}{4\pi} \int \text{div } [E, H]_m d\tau = - \frac{c}{4\pi} \int [E, H]_n dS.$$

Taking the right-hand side as the total energy flow across the surface,  $(K_{ij}E_i \dot{E}_j)/4\pi$  is the time rate of change of the electrical energy density  $(\frac{1}{2} \dot{H}_i^2)/4\pi$  is the time rate of change of the magnetic energy density.

Differentiating with respect to time the expression for  $u$ ,

$$\frac{K_{ij}(E_i \dot{E}_j + E_j \dot{E}_i)}{8\pi} = \dot{u} = \frac{K_{ij}E_i \dot{E}_j}{4\pi}$$

so that

$$K_{ij}(E_i \dot{E}_j - E_j \dot{E}_i) = 0$$

which can be written in the form,

$$K_{ij}E_i \dot{E}_j - K_{ij}E_j \dot{E}_i = 0.$$

The "dummy" suffixes can be transposed in the second term giving,

$$(K_{ij} - K_{ji})E_i \dot{E}_j = 0.$$

Since this expression must hold for any value of the field strength,

$$K_{ij} = K_{ji}$$

i.e., the dielectric tensor  $K_{ij}$  is symmetrical and has only six different components.

### 4. Transformation of $K_{ij}$ into Normal Form

The equation  $K_{ij}x_i x_j = (\text{a constant})$  represents a central conicoid and, since the discriminating cubic equation,

$$|(K_{ij} - \delta_{ij}\lambda)| = 0$$



(where  $\delta_{ij}$  is the substitution tensor) has in general three distinct roots for  $\lambda$ , there are three mutually perpendicular principal directions. These are given by  $l_j \lambda = l_i K_{ij}$ . If the  $x_i$  are measures of the electric field strengths  $E_i$ , then  $K_{ij} E_j$  is  $8\pi u$  and, since the energy is positive for all systems of  $E_i$ , the equation has always a definite form and so must represent an ellipsoid.

Referring the equation to the three mutually perpendicular principal directions  $O\xi_i$  as axes

$$K_{ij} x_i x_j = K' \xi_1^2 + K'' \xi_2^2 + K''' \xi_3^2 = (\text{a constant}).$$

$K'$ ,  $K''$ , and  $K'''$  are called the principal dielectric constants.

In this normal form,

$$D_1 = K' E_1, \text{ etc.}$$

$$8\pi u = K' E_1^2 + K'' E_2^2 + K''' E_3^2 = \frac{D_1^2}{K'} + \frac{D_2^2}{K''} + \frac{D_3^2}{K'''}$$

## 5. Electromagnetic Waves in a Crystalline Dielectric

The differential equations for the  $E_i$  are,

$$\frac{K}{c^2} \frac{\partial^2 E_i}{\partial t^2} = \Delta^2 E_i - \text{grad div } E_i.$$

Taking a solution similar to the isotropic case,

$$E_1 = a' e^{i2\pi(\alpha t - l_j x_j)/\lambda}, \text{ etc.}$$

and substituting these values for  $E_1$ ,  $E_2$  and  $E_3$  in the equations for  $E_i$ , we get, the axes being rectangular

$$\frac{K' \alpha^2 E_1}{c^2} = E_1 - l_1 (l_j E_j),$$

$$\frac{K'' \alpha^2 E_2}{c^2} = E_2 - l_2 (l_j E_j),$$

$$\frac{K''' \alpha^2 E_3}{c^2} = E_3 - l_3 (l_j E_j).$$

If these three equations are simultaneously true then,

$$\frac{l_1^2}{K' \frac{\alpha^2}{c^2} - 1} + \frac{l_2^2}{K'' \frac{\alpha^2}{c^2} - 1} + \frac{l_3^2}{K''' \frac{\alpha^2}{c^2} - 1} + 1 = 0$$

i.e., for every value of  $l_i$  there are in general two values of  $\alpha$  so that two plane waves progress along the same wave normal with velocities  $\alpha$  and  $\alpha'$ . Putting  $c^2/K' = v'^2$ , etc. where  $v'$  will be the velocity along  $Ox_1$ , and rewriting the equation we get,

$$\frac{l_1^2}{\alpha^2 - v'^2} + \frac{l_2^2}{\alpha^2 - v''^2} + \frac{l_3^2}{\alpha^2 - v'''^2} = 0.$$

Or, in terms of the refractive index,

$$\frac{l_1^2}{\frac{1}{n^2} - \frac{1}{K'}} + \frac{l_2^2}{\frac{1}{n^2} - \frac{1}{K''}} + \frac{l_3^2}{\frac{1}{n^2} - \frac{1}{K'''}} = 0.$$

Putting  $x_1 = l_1\alpha$ , etc. we get,

$$\frac{x_1^2}{\alpha^2 - v'^2} + \frac{x_2^2}{\alpha^2 - v''^2} + \frac{x_3^2}{\alpha^2 - v'''^2} = 0.$$

These equations define the wave-normal surface.

## 6. The Relations between $E_i$ , $D_i$ , $H_i$ , and the Wave-normal $l_i$ in a Crystalline Dielectric

The field equations are,

$$\text{curl } H_i - \frac{\dot{D}_i}{c} = 0$$

$$\text{curl } E_i + \frac{\dot{H}_i}{c} = 0.$$

$E_i$  and  $H_i$  are each proportional to  $e^{i2\pi(\alpha t - l_j x_j)/\lambda}$  so that,

$$\begin{aligned} \dot{D}_i &= i \frac{2\pi\alpha}{\lambda} D_i, & \text{curl } E_i &= i \frac{2\pi}{\lambda} [l, E]_i \\ \dot{H}_i &= i \frac{2\pi\alpha}{\lambda} H_i, & \text{curl } H_i &= i \frac{2\pi}{\lambda} [l, H]_i. \end{aligned}$$

Hence

$$i \frac{2\pi}{\lambda} [l, H]_i = i \frac{\alpha 2\pi}{c\lambda} D_i \quad \text{or} \quad \frac{\alpha}{c} D_i = [l, H]_i$$

and

$$i \frac{2\pi}{\lambda} [l, E]_i = -i \frac{\alpha 2\pi}{c\lambda} H_i \quad \text{or} \quad \frac{\alpha}{c} H_i = -[l, E]_i$$

i.e.,  $D_i$  and  $H_i$  are at right-angles to  $l_i$ , and  $E_i$  lies in the plane of  $D_i$  and  $l_i$  but, generally, cannot be at right-angles to  $l_i$  since it does not coincide with  $D_i$ .

Eliminating  $H_i$ , we get,

$$\begin{aligned} \frac{\alpha D_i}{c} &= -\frac{c}{\alpha} [l, [l, E]]_i \\ &= -\{E_i - l_i(l_j E_j)\} \frac{c}{\alpha} \end{aligned}$$

so that

$$D_i = \{E_i - l_i(l_j E_j)\} \frac{c^2}{\alpha^2}.$$



This expression is the general one connecting  $D_i$  and  $E_i$  and in the isotropic case degenerates into  $D_i = n^2 E_i$ .

It can be written,

$$D_1 = \frac{c^2}{\alpha^2} \left\{ \frac{D_1}{K'} - l_1(l_j E_j) \right\} = n^2 \left\{ \frac{D_1}{K'} - l_1(l_j E_j) \right\},$$

or

$$D_1 = - \frac{l_1(l_j E_j)}{\frac{1}{n^2} - \frac{1}{K'}}$$

where  $n$  is the refractive index in the direction  $l_i$ , and two similar equations for  $D_2$  and  $D_3$ .

### 7. Relation between $D_i$ and the Wave-normal $l_i$ .

(a) Let the direction-cosines of  $D_i$  be  $m_i$  and its amplitude  $A$ .  $E_1 = D_1/K'$ , etc. so that in  $E_1 = a' e^{i(2\pi/\lambda)(\alpha t - l_j x_j)}$ , etc.

$$a' = A \frac{m_1}{K'}, \quad \text{etc.}$$

We have,

$$D_i \frac{\alpha^2}{c^2} = E_i - l_i(l_j E_j)$$

so that,

$$E_1 K' \frac{\alpha^2}{c^2} = E_1 - l_1(l_j E_j)$$

and two similar equations.

Hence,

$$m_1 \left( \frac{c^2}{K'} - \alpha^2 \right) = c^2 l_1 \left( \frac{l_1 m_1}{K'} + \text{etc.} \right) = l_1 P \quad \text{say}$$

so that,

$$m_1 = \frac{l_1 P}{\frac{c^2}{K'} - \alpha^2}, \quad m_2 = \text{etc.}$$

For every value of  $l_i$  there are in general two values of  $\alpha$  and  $\alpha'$ , and, thus,

$$m_1 : m_2 : m_3 = \frac{l_1}{\frac{c^2}{K'} - \alpha^2} : \frac{l_2}{\frac{c^2}{K''} - \alpha^2} : \frac{l_3}{\frac{c^2}{K'''} - \alpha^2}$$

$$m_1' : m_2' : m_3' = \frac{l_1}{\frac{c^2}{K'} - \alpha'^2} : \frac{l_2}{\frac{c^2}{K''} - \alpha'^2} : \frac{l_3}{\frac{c^2}{K'''} - \alpha'^2}.$$

Hence  $m_i m_i'$  is proportional to,

$$\frac{l_1^2}{\left(\frac{c^2}{K'} - \alpha^2\right)\left(\frac{c^2}{K'} - \alpha'^2\right)} + \text{etc.}$$

$$= \frac{1}{\alpha^2 - \alpha'^2} \left\{ \frac{l_1^2}{\frac{c^2}{K'} - \alpha^2} + \text{etc.} - \left( \frac{l_1^2}{\frac{c^2}{K'} - \alpha'^2} + \text{etc.} \right) \right\} = 0$$

so that  $m_i$  and  $m_i'$  are at right-angles, i.e., the two displacements for the wave-normal  $l_i$  are at right-angles.

(b) We have from (5),

$$-\frac{l_1^2}{\frac{c^2}{K'} - \alpha^2} + \frac{l_2^2}{\frac{c^2}{K''} - \alpha^2} + \frac{l_3^2}{\frac{c^2}{K'''} - \alpha^2} = 0$$

and thus from (a),

$$\left(\frac{c^2}{K'} - \alpha^2\right) m_1^2 + \left(\frac{c^2}{K''} - \alpha^2\right) m_2^2 + \left(\frac{c^2}{K'''} - \alpha^2\right) m_3^2 = 0$$

so that,

$$\alpha^2 = \frac{c^2 m_1^2}{K'} + \frac{c^2 m_2^2}{K''} + \frac{c^2 m_3^2}{K'''}$$

and there is thus only one wave velocity corresponding to each direction of the displacement vector.

(c) From (a),

$$m_1 = \frac{l_1 \xi}{v'^2 - \alpha^2},$$

where

$$g^2 = \frac{1}{\sum \frac{l_i^2}{(v'^2 - \alpha^2)^2}}$$

and two similar equations for  $m_2$ ,  $m_3$ .

## 8. Energy of Electrical and Magnetic Vibrations

We have,

$$u = \frac{E_i D_i}{8\pi} = \frac{c^2 \{E_i^2 - (E_i l_i)^2\}}{8\pi \alpha^2}$$

and

$$t = \frac{H_i^2}{8\pi} = \frac{c^2 [E, l]_i^2}{8\pi c v^2} = \frac{c^2 \{E_i^2 l_i^2 - (E_i l_i)^2\}}{8\pi c v^2}$$

so that,

$$w = u + t = 2u = 2l = \frac{n^2 \{E_i^2 - (E_i l_i)^2\}}{4\pi}.$$

9. Relations between  $E_i$ ,  $D_i$ ,  $l_i$ ,  $w$  and  $n$

By (6) we have

$$D_i = n^2 \{E_i - l_i (E_j l_j)\}$$

so that,

$$D_i^2 = n^4 \{E_i^2 - (E_j l_j)^2\} = 4\pi n^2 w$$

and

$$n^2 = \frac{D_i^2}{4\pi w} = \frac{D_i^2}{E_i D_i}.$$

Thus

$$\begin{aligned} l_i &= \frac{E_i - \frac{D_i}{n^2}}{E_j l_j} = \frac{E_i - \frac{D_i}{n^2}}{\sqrt{E_j^2 - \frac{D_j^2}{n^4}}} \\ &= \frac{E_i - \frac{(E_j D_j) D_i}{D_j^2}}{\sqrt{E_j^2 - \frac{(E_j D_j)^2}{D_j^2}}} = \frac{E_i D_j^2 - D_i (E_j D_j)}{\sqrt{D_j^2 \{E_j^2 D_j^2 - (E_j D_j)^2\}}}. \end{aligned}$$

10. The Energy Flow  $N_i$  in a Crystalline Dielectric—the Ray

We have from (3),  $N_i = (c/4\pi) [E, H]_i$  and hence the energy flow diverges from the wave-normal at an angle  $\theta$  say. This path of flow of the energy is called the ray and is at right-angles to  $E_i$  and  $H_i$ , and lies in the plane of  $l_i$ ,  $E_i$ , and  $D_i$ . Inserting the values for  $H_i$  we get,

$$N_i = \frac{cn}{4\pi} [E, [E, l]]_i = \frac{cn}{4\pi} \{l_i E_j^2 - E_i (E_j l_j)\}.$$

Hence,

$$\begin{aligned} N_i^2 &= \frac{c^2 n^2}{(4\pi)^2} \{(E_j^2)^2 - E_i^2 (E_j l_j)^2\} \\ &= \frac{c^2 n^2}{(4\pi)^2} E_i^2 \{E_j^2 - (E_j l_j)^2\} = \frac{c^2 E_i^2 w}{4\pi}. \end{aligned}$$

Also,

$$|N_i| \cos \theta = N_i l_i = \frac{cn}{4\pi} l_i \{l_i E_j^2 - E_i (E_j l_j)\} = \frac{cw}{n} \quad \text{by (8).}$$

Again,



$$\begin{aligned}
N_i D_i &= \frac{cn}{4\pi} \{l_i E_j^2 - E_i(E_j l_i)\} D_i \\
&= \frac{cn^3}{4\pi} \{l_i E_j^2 - E_i(E_j l_i)\} \{E_i - l_i(E_j l_i)\} && \text{by (6)} \\
&= -\frac{cn^3}{4\pi} \{E_j^2 - (E_j l_i)^2\} E_i l_i \\
&= -ncE_j l_i w && \text{by (8).}
\end{aligned}$$

### 11. The Ray Index, $r$

$N_i$  is the amount of energy crossing unit surface normally, in unit time. Imagine a cylinder erected on this base of unit area, with its length parallel to  $p_i$ , the direction of  $N_i$ , and of height  $\beta$  where  $\beta$  is the velocity along  $p_i$ . If the energy density within it is  $w$  then in unit time an amount of energy  $\beta w$  will pass through the unit area and thus,

$$|N_i| = \beta w.$$

We have,

$$N_i l_i = \frac{cw}{n}.$$

So that

$$\beta = \frac{c}{n(p_i l_i)} = \frac{\alpha}{p_i l_i}$$

i.e., the wave velocity is the projection of the ray velocity on the wave-normal.

Also, since

$$\begin{aligned}
N_i^2 &= \frac{c^2 E_i^2 w}{4\pi} \\
\beta^2 &= \frac{N_i^2}{w^2} = \frac{c^2}{4\pi w} E_i^2.
\end{aligned}$$

We define the ray index,  $r$ , by  $r = c/\beta$  so that,

$$\begin{aligned}
r^2 &= \left(\frac{c}{\beta}\right)^2 = \frac{c^2 4\pi w}{c^2 E_i^2} = \frac{E_i D_i}{E_i^2} \\
&\left(\text{cf. } n^2 = \frac{D_i^2}{E_i D_i}\right).
\end{aligned}$$

Again, inserting the value for  $l_i$  in the expression for  $N_i$ , we get,

$$\begin{aligned}
N_i &= \frac{cn(E_j D_j \{E_i(E_j D_j) - D_i E_j^2\})}{4\pi \sqrt{D_j^2 \{E_i^2 D_j^2 - (E_j D_j)^2\}}} \\
&= \frac{c \sqrt{E_j D_j} \{E_i(E_j D_j) - D_i E_j^2\}}{4\pi \sqrt{E_j^2 D_j^2 - (E_j D_j)^2}}.
\end{aligned}$$

Since  $p_i = N_i / |N_i|$  and as,

$$|N_i| = \frac{c}{4\pi} \sqrt{(E_j D_j) E_j^2}$$

$$-p_i = \frac{D_i E_j^2 - E_i (E_j D_j)}{\sqrt{E_j^2 \{E_j^2 D_j^2 - (E_j D_j)^2\}}}$$

$$\left( \text{cf. } l_i = \frac{E_i D_j^2 - D_i (E_j D_j)}{\sqrt{D_j^2 \{E_j^2 D_j^2 - (E_j D_j)^2\}}} \right).$$

## 12. Relations of Wave and Ray Vectors

We have,

$$E_i p_i = 0; \quad D_i l_i = 0; \quad p_i l_i = \cos \theta; \quad r = n \cos \theta$$

where  $\theta$  is the angle between the wave-normal and the ray.

Hence in the equation

$$D_i = n^2 \{E_i - l_i (E_i l_i)\} \quad \text{of (6)}$$

$$D_i p_i = n^2 \{E_i p_i - p_i l_i (E_i l_i)\} = -n^2 (E_i l_i) \cos \theta$$

or

$$E_i l_i = -\frac{D_i p_i}{n^2 \cos \theta}.$$

Since  $E_i$ ,  $D_i$  and  $p_i$  are coplanar,

$$p_i = a D_i + b E_i$$

where  $a$  and  $b$  are constants.

Hence

$$p_i^2 = a p_i D_i + b p_i E_i \quad \text{i.e.,} \quad a p_i D_i = 1$$

and

$$p_i l_i = a l_i D_i + b l_i E_i \quad \text{i.e.,} \quad b l_i E_i = \cos \theta.$$

So that,

$$p_i = \frac{D_i}{p_i D_i} + \frac{E_i \cos \theta}{l_i E_i}$$

and

$$E_i = \frac{l_i E_i}{\cos \theta} \left\{ p_i - \frac{D_i}{p_i D_i} \right\}.$$

Inserting the value for  $E_j l_j$  we get,

$$E_i = \frac{-D_i p_i \left( p_i - \frac{D_i}{p_i D_i} \right)}{n^2 \cos^2 \theta} = \frac{D_i - p_i (D_i p_i)}{r^2}.$$

Taking all these wave and ray equations together and comparing them, we see that they have the following correspondence in their terms;

$$\begin{array}{cccccccccccccccc}
 E_i & D_i & l_i & p_i & \alpha & n & K' & K'' & K''' & v' & v'' & v''' & c & \text{wave equations} \\
 D_i & E_i & -p_i & -l_i & \frac{1}{\beta} & \frac{1}{r} & \frac{1}{K'} & \frac{1}{K''} & \frac{1}{K'''} & \frac{1}{v'} & \frac{1}{v''} & \frac{1}{v'''} & \frac{1}{c} & \text{ray equations}
 \end{array}$$

### 13. The Ray Equations

We have for the wave, from (6),

$$D_1 = \frac{-l_1(l_i E_i)}{\frac{1}{n^2} - \frac{1}{K'}} = \frac{-c^2 l_1(l_i E_i)}{\alpha^2 - v'^2}$$

and two similar equations for  $D_2$  and  $D_3$  and also, from (5),

$$\frac{l_1^2}{\alpha^2 - v'^2} + \frac{l_2^2}{\alpha^2 - v''^2} + \frac{l_3^2}{\alpha^2 - v'''^2} = 0.$$

Substituting the corresponding terms from (12) we get,

$$E_1 = \frac{-p_1(p_i D_i)}{c^2 \left( \frac{1}{\beta^2} - \frac{1}{v'^2} \right)}$$

and two similar equations for  $E_2$  and  $E_3$ .

Also,

$$\frac{p_1^2}{\frac{1}{\beta^2} - \frac{1}{v'^2}} + \frac{p_2^2}{\frac{1}{\beta^2} - \frac{1}{v''^2}} + \frac{p_3^2}{\frac{1}{\beta^2} - \frac{1}{v'''^2}} = 0$$

and

$$\frac{p_1^2}{r^2 - K'} + \frac{p_2^2}{r^2 - K''} + \frac{p_3^2}{r^2 - K'''} = 0.$$

These equations define the ray surface which, if we put  $x_i = \beta p_i$ , may be written,

$$\frac{v'^2 x_1^2}{v'^2 - \beta^2} + \frac{v''^2 x_2^2}{v''^2 - \beta^2} + \frac{v'''^2 x_3^2}{v'''^2 - \beta^2} = 0.$$

Thus to every value of  $p_i$  there are two values of  $\beta$  or  $r$ .

### 14. Relations of $l_i$ and $p_i$

In any actual case only one of the vectors  $l_i$  and  $p_i$  is given and the other must be calculated from it. Knowing the  $l_i$  or  $p_i$ , the  $D_i$  and  $E_i$  can be calculated from the equations of (6) and (13).

(a) We have,

$$D_1 = E_1 K' = \frac{-p_1(D_i p_i) \beta^2}{v'^2 - \beta^2}$$

and two similar equations for  $D_2$  and  $D_3$ .



Also,

$$D_i p_i = - (E_i l_i) n^2 \cos \theta = \frac{c^2 (E_i l_i)}{\alpha \beta}.$$

Thus,

$$\frac{l_1 \alpha}{\alpha^2 - v'^2} = \frac{p_1 \beta}{\beta^2 - v'^2}$$

and two similar equations for  $l_2, l_3, p_2, p_3$ .

(b) We have already, from (5), the relations between the direction-cosines of the wave-normals so that we derive,

$$\frac{l_1 p_1}{\beta^2 - v'^2} + \frac{l_2 p_2}{\beta^2 - v'^2} + \frac{l_3 p_3}{\beta^2 - v'^2} = 0.$$

(c) Again, from (a) we can write,

$$\beta p_1 - \alpha l_1 = \frac{l_1 \alpha (\beta^2 - \alpha^2)}{\alpha^2 - v'^2}$$

and two similar equations.

Squaring and adding these three equations, we have,

$$\alpha^2 (\beta^2 - \alpha^2) = \frac{1}{\sum \left\{ \frac{l_i}{\alpha^2 - v'^2} \right\}^2} = g^2 \quad \text{by (7)}$$

$\alpha$  is already known by the wave equation in terms of  $l_i$  and therefore  $\alpha^2 (\beta^2 - \alpha^2)$ , and thus  $\beta$ , can be expressed in terms of  $l_i$ . Hence  $p_i$  is expressed as a function of  $l_i$ .

We can write,

$$p_1 = \frac{l_1}{\alpha \beta} \left\{ \alpha^2 + \frac{g^2}{\alpha^2 - v'^2} \right\}$$

and two similar equations for  $p_2, p_3, l_2, l_3$ .

Hence, since to each value of  $l_i$  there are two values of  $\alpha$ , there must in general be two rays  $p_i$  for each  $l_i$ .

(d) By the relations of (12) we can write,

$$l_1 = \alpha \beta p_1 \left\{ \frac{1}{\beta^2} + \frac{g^2}{\frac{1}{\beta^2} - \frac{1}{v'^2}} \right\}$$

and two similar equations for  $l_2, l_3, p_2, p_3$ .

Hence, since to each value of  $p_i$  there are two values of  $\beta$ , there must in general be two wave-normals  $l_i$  for each  $p_i$ .

## 15. Relations of the Wave-normal Surface and the Ray Surface

A small change in either the electrical intensity or the displacement

will bring about a small change in the  $l_i$  relative to the  $p_i$ . We now investigate this relation.

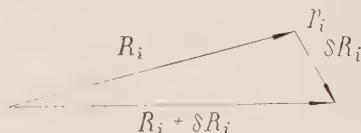


FIG. 1. The direction of the tangent plane to the ray surface.

Let  $p_i$  be the ray direction and  $\beta$  the ray velocity in this direction. If  $R_i = \beta p_i$  then the end of  $R_i$  sweeps out the ray surface. Let  $l_i$  be the wave-normal associated with  $p_i$  according to (14).

Since  $p_i c = r R_i$ , we have from (12),

$$c^2 E_i = (R_i^2) D_i - R_i (D_i R_i).$$

If the quantities be considered as functions of a variable  $t$ , say, then the displacement of  $R_i$  will be  $\delta R_i = \dot{R}_i \delta t$  where the dot indicates differentiation with respect to  $t$ .

We have,

$$c^2 \dot{E}_i = \dot{D}_i (R_i^2) + 2 D_i (R_i \dot{R}_i) - \dot{R}_i (D_i R_i) - R_i (D_i \dot{R}_i) - R_i (\dot{D}_i R_i).$$

Multiplying these equations by the appropriate  $D_i$  and adding, we have,

$$\begin{aligned} c^2 \dot{E}_i D_i &= (\dot{D}_i D_i) (R_i^2) - (D_i R_i) (\dot{D}_i R_i) + 2 \{ (D_i^2) (\dot{R}_i R_i) - (R_i D_i) (\dot{R}_i D_i) \} \\ &= \dot{D}_i \{ (R_i^2) D_i - R_i (D_i R_i) \}_i + 2 \dot{R}_i \{ (D_i^2) R_i - D_i (R_i D_i) \}_i \\ &= c^2 \dot{D}_i E_i + 2 \dot{R}_i [[D, R], D]_i \end{aligned}$$

i.e.,  $2 \dot{R}_i [[D, R], D]_i = 0$  since  $D_i \dot{E}_i = E_i \dot{D}_i$ .

The vector  $[[D, R], D]_i$  is perpendicular to the normal to the plane of  $D_i$  and  $p_i$  and is also at right-angles to  $D_i$ . It is therefore parallel to  $l_i$ . We have then  $\dot{R}_i [[D, R], D]_i = 0$  and hence the displacement of  $R_i$ , being  $\delta R_i = \dot{R}_i \delta t$ , must be at right-angles to  $l_i$  since  $l_i \delta R_i = 0$ . That is, the tangent plane to the ray surface at the end of a radius-vector is always at right-angles to the corresponding wave-normal.

The principal axes of the wave-normal surface and the ray surface coincide and therefore the wave-normal surface is the pedal surface of the ray surface and conversely the ray surface is the envelope of the planes at right-angles to the radii-vectores of the wave-normal surface.

## 16. Derivation of the Wave-normal Ellipsoid<sup>4</sup>

By (5) we have for the relation between the wave-normal  $l_i$  and the

<sup>4</sup> This surface is also called the indicatrix (Fletcher), the indexellipsoid (Pockels and Szivessy), the normalenellipsoid (Born). As the term "indicatrix" has long had a definite

velocities  $\alpha$  of the two waves propagated along it,

$$\frac{l_1^2}{\alpha^2 - v'^2} + \frac{l_2^2}{\alpha^2 - v''^2} + \frac{l_3^2}{\alpha^2 - v'''^2} = 0.$$

Hence if waves travel outwards from a point within the crystal in all directions, the limits of their travel after unit time along the normals will form a twofold surface, the wave-normal surface, which is of the fourth degree. Such a surface is a complicated one and it is more convenient to take as reference an ellipsoid derived from the energy equation (4),

$$\frac{D_1^2}{K'} + \frac{D_2^2}{K''} + \frac{D_3^2}{K'''} = 8\pi u.$$

Taking the  $x_i$  as measures of the  $D_i$  and with suitable adjustments we can put,

$$\frac{x_1^2}{K'} + \frac{x_2^2}{K''} + \frac{x_3^2}{K'''} = 1, \quad \text{or} \quad \frac{x_1^2}{n'^2} + \frac{x_2^2}{n''^2} + \frac{x_3^2}{n'''^2} = 1.$$

This is an ellipsoid whose principal axes coincide with the dielectric axes and are proportional to the roots of the principal dielectric constants or to the principal refractive indices. It is called here the wave-normal ellipsoid. By reference to it the course of the propagation of light in crystals can be illustrated and examined in the following manner.

#### 17. Refractive Indices for the Wave-normal $l_i$

Let a radius-vector of the ellipsoid represent a wave-normal  $l_i$ . Then, by (6),  $D_i$ , which we shall take as the "vibration," must lie in a plane at right-angles to this radius vector. Let  $l_i x_i = 0$  be such a plane through the origin. It will cut the ellipsoid in an ellipse and the principal axes of the ellipse give in direction and magnitude the two values of  $D_i$  demanded by electromagnetic theory. That this is so we prove as follows.

For the radius-vector of length  $r$ ,

$$r^2 = x_i^2 = f(x_i) \quad \text{say.}$$

We have therefore to find the maximum and minimum values for  $r$  having regard to the conditions,

$$0 = \frac{x_1^2}{K'} + \frac{x_2^2}{K''} + \frac{x_3^2}{K'''} - 1 = \phi(x_i) \quad \text{say,}$$

and

$$0 = l_i x_i = \psi(x_i) \quad \text{say.}$$

meaning in the geometry of higher surfaces and curves and as the expression "indexellipsoid" is ambiguous, Born's name is probably best. "Wave-normal ellipsoid" is used here to make the reference as precise as possible.



Forming  $df + \lambda d\phi + 2 \mu d\psi$  where  $\lambda$  and  $2 \mu$  are undetermined multipliers, and equating the coefficients of the  $dx_i$  to zero,

$$x_1 + \frac{\lambda x_1}{K'} + \mu l_1 = 0, \quad x_2 + \frac{\lambda x_2}{K''} + \mu l_2 = 0, \quad x_3 + \frac{\lambda x_3}{K'''} + \mu l_3 = 0.$$

The values of  $x_i$  which satisfy these equations are those that determine the turning values of  $r^2$ . Multiplying the equations by the  $x_i$  and adding gives  $r^2 = -\lambda$  and this gives on substitution,

$$x_1 = \frac{\mu l_1}{\frac{r^2}{K'} - 1}, \quad x_2 = \frac{\mu l_2}{\frac{r^2}{K''} - 1}, \quad x_3 = \frac{\mu l_3}{\frac{r^2}{K'''} - 1}.$$

Inserting these values in  $l_i x_i = 0$ ,

$$\frac{l_1^2}{\frac{r^2}{K'} - 1} + \frac{l_2^2}{\frac{r^2}{K''} - 1} + \frac{l_3^2}{\frac{r^2}{K'''} - 1} = 0$$

which gives two solutions for  $r^2$ , the turning values.

By (5) this is the equation which defines the refractive indices of the two waves proceeding along the wave-normal  $l_i$  so that the lengths of the major and minor axes of the elliptic section give the refractive indices of the waves propagated along the radius-vector  $l_i$ .

Again, if we multiply the equations by the  $l_i$  and add, we get,

$$\lambda \left( \frac{x_1 l_1}{K'} + \frac{x_2 l_2}{K''} + \frac{x_3 l_3}{K'''} \right) + \mu = 0$$

which gives then, as the three equations defining the  $x_i$  for the turning values of  $r^2$ ,

$$x_1 - \frac{r^2 x_1}{K'} + l_1 r^2 \left( \frac{x_1 l_1}{K'} + \frac{x_2 l_2}{K''} + \frac{x_3 l_3}{K'''} \right) = 0$$

and two similar equations. If in these equations the  $x_i$  are replaced by the  $D_i$  and  $x_i/K'$ , etc. by  $E_i$ , etc., then,

$$D_1 = n^2 \left( \frac{D_1}{K'} - l_1 (E_i l_i) \right)$$

and two similar equations, and these by (6) define the electrical displacements associated with the wave-normal  $l_i$ .

## 18. Vibration Directions for the Wave-normal $l_i$

From the values obtained in (17) for the  $x_i$  the ratios of the direction-cosines of the axes of the elliptic section at right-angles to  $l_i$  are,

$$\frac{l_1}{v'^2 - \alpha^2} : \frac{l_2}{v''^2 - \alpha^2} : \frac{l_3}{v'''^2 - \alpha^2}$$

which are the values determined for the direction-cosines  $m_i$  of  $D_i$  in (7). We note further that these electrical displacements are at right-angles as required by (7).

### 19. Ray Direction and Ray Index for the Wave-normal $l_i$

Let  $x_i'$  be the end of one of the principal axes of the elliptic section at

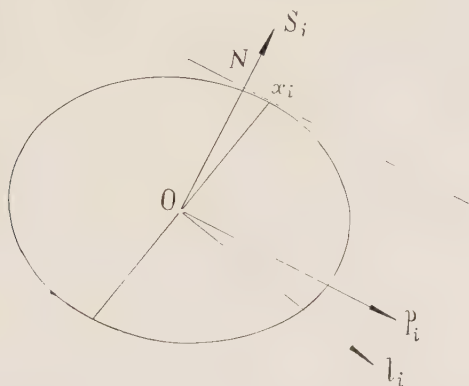


FIG. 2. Section of the wave-normal ellipsoid containing the wave-normal  $l_i$  and one of the principal axes  $Ox_i$  of the elliptic section at right-angles to it.

right-angles to the wave-normal  $l_i$ . This axis defines one value of  $D_i$  in magnitude and direction, i.e., on the appropriate scale,  $x_i' = D_i$ . By (6),

$$D_i = \frac{l_i(l_i E_i)}{\frac{1}{n'^2} - \frac{1}{n^2}}$$

so that,

$$n = |D_i| = \left[ \sum \left\{ \frac{l_i(l_i E_i)}{\frac{1}{n'^2} - \frac{1}{n^2}} \right\}^2 \right]^{1/2}.$$

By (14)

$$g^2 = \frac{1}{\sum \left( \frac{l_i}{\alpha^2 - v'^2} \right)^2} = \frac{c^4}{\sum \frac{l_i^2}{\left( \frac{1}{n'^2} - \frac{1}{n^2} \right)^2}}.$$

Hence,

$$l_i E_i = \frac{ng}{c^2}.$$

We have thus for the co-ordinates  $x_i'$ ,

$$x_1' = \frac{l_1 n g}{c^2 \left( \frac{1}{n'^2} - \frac{1}{n^2} \right)}$$

and similar equations for  $x_2'$  and  $x_3'$ .

The tangent to the elliptic section at  $x_i'$  is at right-angles to  $Ox_i'$  and is thus perpendicular to the plane of  $l_i$  and  $Ox_i'$  since it also lies in the central plane at right-angles to  $l_i$ . Draw the tangent plane to the ellipsoid at  $x_i'$  and let  $ON$ , with direction-cosines  $s_i$ , be the normal to this plane from the origin. Since  $ON$  is also at right angles to the line through  $N$  parallel to the tangent to the elliptic section at  $x_i'$ , it must be co-planar with  $l_i$  and  $Ox_i'$ .

We have,

$$\begin{aligned} s_1 &= \frac{\frac{x_1'}{n'^2}}{\left\{ \sum \left( \frac{x_i}{n'^2} \right)^2 \right\}^{1/2}} = \frac{x_1' q}{n'^2}, \quad \text{say,} \\ &= \frac{l_1 n g q}{c^2 n'^2 \left( \frac{1}{n'^2} - \frac{1}{n^2} \right)} \end{aligned}$$

and similar equations for  $s_2$  and  $s_3$ .

By (14) we have, for the ray corresponding to the wave-normal  $l_i$ ,

$$p_1 = \frac{r l_1}{n} \left\{ 1 - \frac{n^2 g^2}{c^4 \left( \frac{1}{n'^2} - \frac{1}{n^2} \right)} \right\}$$

and similar equations for  $p_2$  and  $p_3$ .

Hence,

$$\begin{aligned} s_i p_i &= \frac{g q r}{c^2} \sum \left[ \frac{l_i^2}{n'^2 \left( \frac{1}{n'^2} - \frac{1}{n^2} \right)} \left\{ 1 - \frac{n^2 g^2}{c^4 \left( \frac{1}{n'^2} - \frac{1}{n^2} \right)} \right\} \right] \\ &= \frac{g q r}{c^2} \left\{ \sum \frac{l_i^2}{n'^2 \left( \frac{1}{n'^2} - \frac{1}{n^2} \right)} - \frac{g^2}{c^4} \sum \frac{l_i^2 n^2}{n'^2 \left( \frac{1}{n'^2} - \frac{1}{n^2} \right)^2} \right\}. \end{aligned}$$

From (5)

$$\sum \frac{l_i^2}{n'^2 \left( \frac{1}{n'^2} - \frac{1}{n^2} \right)} = 1$$

also,

$$\sum \frac{l_i^2}{\left( \frac{1}{n'^2} - \frac{1}{n^2} \right)} = 0$$



so that,

$$\sum \frac{l_1^2}{n'^2 \left( \frac{1}{n'^2} - \frac{1}{n^2} \right)^2} = \sum \frac{l_1^2}{n^2 \left( \frac{1}{n'^2} - \frac{1}{n^2} \right)^2}$$

$$\therefore s_i p_i = 0$$

i.e.  $p_i$  is given by the two conditions, (a), it lies in the plane of  $l_i$ ,  $Ox_i'$  and  $ON$ , and, (b), it is at right-angles to  $ON$ .

Further,  $ON = n \cos \theta$  and hence is equal to the ray index  $r$ . We note also that, from the geometry of the ellipsoid,  $ON$  will be a principal semi-diameter of the cross section of the cylinder with axis  $p_i$  and tangential to the ellipsoid along its intersection with the plane diametral to  $p_i$ .

These relations being established between the wave-normal ellipsoid and the electromagnetic vectors, all of the remaining relations depend only on the geometry of the ellipsoid itself. Beer, Becke and Wright<sup>5</sup> have pointed out that the full optical relations can be developed from the wave-normal ellipsoid by consideration of the cones defined by the intersection of the ellipsoid with a sphere of variable radius corresponding to the refractive index. In concluding this note it is worth drawing attention to the role played in this matter by the focal lines of these cones.

If the wave-normal ellipsoid is

$$\sum \frac{x_i^2}{n'^2} = 0$$

the radii-vectors of length corresponding to the variable refractive index  $r$  give the "equivibration" cone

$$\sum x_1^2 \left( \frac{1}{n'^2} - \frac{1}{r^2} \right) = 0$$

and this degenerates into the two planes of cyclic section of the ellipsoid for  $r = n''$ . Since the coefficients of  $x_1^2$  etc. in the equations to the cone and the ellipsoid differ only by a constant term, the directions of the circular sections are the same in each. The real focal lines of these equivibration cones are given by,

$$x_1^2 \frac{(r^2 - n''^2)}{(n''^2 - n'^2)} - x_2^2 \frac{(n'''^2 - r^2)}{(n'''^2 - n'^2)} = 0 \quad \text{for } n'' < r < n'''$$

and

$$x_1^2 \frac{(r^2 - n'^2)}{(n'''^2 - n'^2)} - x_2^2 \frac{(n'''^2 - r^2)}{(n'''^2 - n'^2)} = 0 \quad \text{for } n' < r < n'''$$

<sup>5</sup> Beer, A.: *Grunert's Arch.*, Th. 16, 223-229 (1851).

Becke, F.: *Tschermak's Min. u. Pet. Mitt.*, **24**, 1-34 (1905).

Wright, F. E.: *Jour. Opt. Soc. Am.*, **7**, 779-817 (1923).

i.e. the cones fall into two sets, the cyclic sections of the ellipsoid forming the boundary between them.

We note in passing that a property of the focal lines of such cones is that the section of the cone by any plane at right-angles to one of them is a conic having for focus the point where the focal line meets the plane. Further, the directrix of the cone corresponding to the point on the focal line lies in the plane of section and is at right-angles to the plane of the focal lines. It is also the directrix of the conic section.

A tangent plane to the equivibration cone along a generator,  $l_i$ , say, is

$$\sum l_i \left( \frac{1}{n'^2} - \frac{1}{r^2} \right) x_i = 0.$$

It is a diametral plane of the ellipsoid and the generator, being a radius of the sphere and thus at right-angles to the curve of its intersection with the ellipsoid, is a principal axis of the elliptic section. Hence the normals to the tangent planes of the equivibration cone give the directions of propagation for which one refractive index is  $r$ . They form the cone

$$\sum \frac{x_i^2}{\left( \frac{1}{n'^2} - \frac{1}{r^2} \right)} = 0,$$

the equirefringence cone, which is reciprocal to the equivibration cone. The relations of two such reciprocal cones are shown in Fig. 3. The focal lines of each cone are at right-angles to the circular sections of the other and the normal plane common to the two cones namely,

$$\sum \frac{\left( \frac{1}{n'''^2} - \frac{1}{n''^2} \right)}{l_i} x_i = 0,$$

bisects the angle between the planes through the generator and the focal lines in each case. The direction-cosines of the generator of the equirefringence cone corresponding to the vibration direction  $l_i$  are in the ratio,

$$\begin{aligned} l_1 \left\{ \left( \frac{1}{n'''^2} - \frac{1}{n'^2} \right) l_3^2 - \left( \frac{1}{n'^2} - \frac{1}{n''^2} \right) l_2^2 \right\} \\ : l_2 \left\{ \left( \frac{1}{n'^2} - \frac{1}{n''^2} \right) l_1^2 - \left( \frac{1}{n'''^2} - \frac{1}{n''^2} \right) l_3^2 \right\} \\ : l_3 \left\{ \left( \frac{1}{n''^2} - \frac{1}{n'''^2} \right) l_2^2 - \left( \frac{1}{n'''^2} - \frac{1}{n'^2} \right) l_1^2 \right\}. \end{aligned}$$

The focal lines of the equirefringence cones are therefore at right-angles to the cyclic planes of the equivibration cones and of the ellipsoid and are thus the same for all. They constitute the wave-normal axes and the

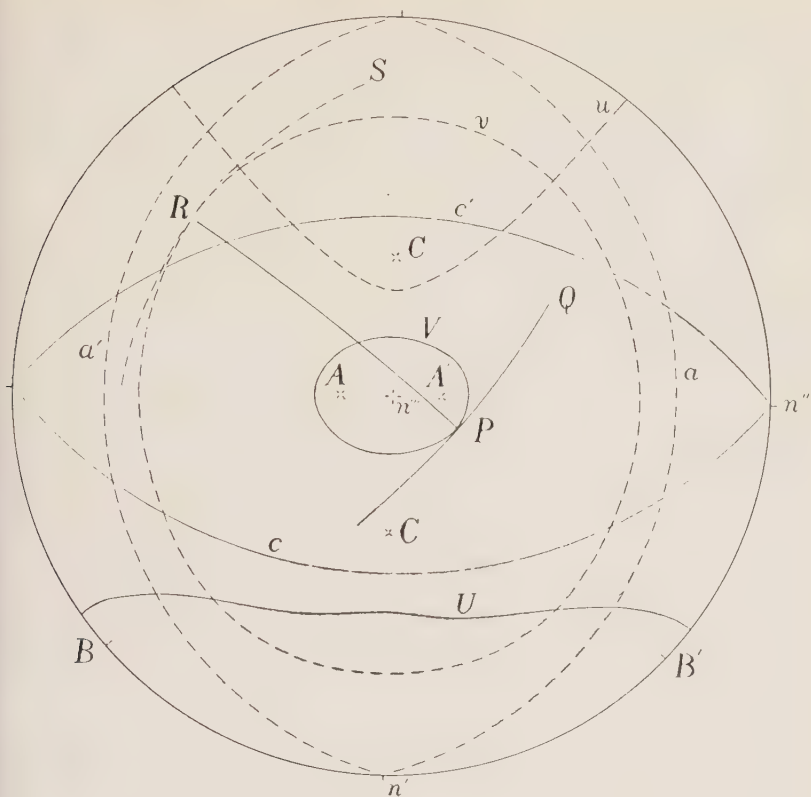


FIG. 3. Reciprocal cones in stereographic projection.  $V$  is a cone of the set  $n'' < r < n'''$  with focal lines at  $A$  and  $A'$ , and  $U$ , focal lines at  $B$  and  $B'$ , one of the set  $n' < r < n''$ .  $c$  and  $c'$  are the planes of circular section,  $r = n''$ . The cone reciprocal to  $V$  is  $v$  and its focal lines are  $C$  and  $C'$  at right-angles to the cyclic sections  $c$  and  $c'$ . The normal plane common to the two cones is  $RP$  and it bisects, in each cone, the angle between the planes through the generator and the focal lines. The circular sections of  $v$  are  $a$  and  $a'$  at right-angles to the focal lines  $A$  and  $A'$  of  $V$ .  $RS$  is the tangent plane to  $v$  at right angles to  $OP$  and  $PQ$  the tangent plane to  $V$  at right-angles to  $OR$ .

refractive indices for the crystal are thus given by cones set about these optic axes. These fall into two sets corresponding to the two sets of equivibration cones as shown in Fig. 3, i.e., (i) for  $n'' < r < n'''$  the cones lie between the principal section  $n'n''$  and the wave-normal axes. Their circular sections, being at right-angles to the focal lines of the reciprocal, equivibration cones, are perpendicular to the plane  $n''n'''$ . (ii) for  $n' < r < n''$  they lie between the principal section  $n''n'''$  and the wave-normal axes, their circular sections being at right-angles to the plane of  $n'$  and  $n''$ . Since they have the same focal lines, these two sets of equire-

fringe cones intersect, thus determining the refractive indices for the direction of propagation defined by the common generators.

Since we are concerned only with directions, it is sufficient to consider the intersection of these equivibration and equirefringence cones with a sphere of reference in which they depict for the crystal the refractive indices and the directions of vibration.<sup>6</sup> Since for a second degree cone the sum of the angles between any generator and the focal lines is a constant, namely the angle between the generators in the plane of the focal lines, the curves of intersection of these cones with the sphere are analogous to plane ellipses.

The directions of vibration for a generator defining a propagation direction is given by the intersection of the normal plane through the generator with the plane at right-angles to the generator. This plane, as we have seen, bisects the angle between the planes containing the generator and the focal lines. If the direction-cosines of the generator are  $l_i$  the direction-cosines of the vibration direction are in the ratio,

$$\begin{aligned} & l_1 \left\{ \left( \frac{1}{n'^2} - \frac{1}{r^2} \right) \left( \frac{1}{n'^2} - \frac{1}{n''^2} \right) l_3^2 - \frac{1}{n''^2} - \frac{1}{r^2} \left( \frac{1}{n'^2} - \frac{1}{n''^2} \right) l_2^2 \right\} \\ & : l_2 \left\{ \left( \frac{1}{n'^2} - \frac{1}{r^2} \right) \left( \frac{1}{n'^2} - \frac{1}{n''^2} \right) l_1^2 - \left( \frac{1}{n'^2} - \frac{1}{r^2} \right) \left( \frac{1}{n'^2} - \frac{1}{n''^2} \right) l_3^2 \right\} \\ & : l_3 \left\{ \left( \frac{1}{n'^2} - \frac{1}{r^2} \right) \left( \frac{1}{n'^2} - \frac{1}{n''^2} \right) l_2^2 - \left( \frac{1}{n'^2} - \frac{1}{r^2} \right) \left( \frac{1}{n'^2} - \frac{1}{n''^2} \right) l_1^2 \right\} \end{aligned}$$

<sup>6</sup> See Johannsen, A.: *Manual of Petrographic Methods*, New York, (1918), pp. 429-434, and Wright, F. E.: *op. cit.*, pp. 790-792 for diagrams of projections of these curves of intersection on the principal planes. Both of these works give a summary of the development of this method of analysis of the optical properties of crystals.



# HIGH-TEMPERATURE PHASES FROM KAOLINITE AND HALLOYSITE\*

HERBERT D. GLASS†

## ABSTRACT

A selected group of kaolinite and halloysite clays of varying structure and impurities are examined by differential thermal and x-ray diffraction methods to establish the significance of the various observed thermal effects. The heating range for the thermal curves extends to 1350° C. and the study of specimens is supplemented by the firing of materials to intermediate temperatures.

Interpretations are given for the cause of the various thermal peaks, and the reasons for lack of agreement in the literature are discussed. The complete firing sequence for kaolinite and halloysite is described.

## INTRODUCTION

Kaolinite-type clays are so widely used in the manufacture of ceramics that many investigators have studied the sequence of phase changes in kaolinite during its firing. The investigators have differed, however, regarding the phase changes observed and also their interpretations as to cause of the thermal reactions. The author therefore examined phase changes in kaolinite and halloysite under controlled firing conditions.

The results of this study show that the disagreements in previous reports may be accounted for by the fact that materials of different degrees of crystallinity, morphology, and purity show different sequences of phase changes at high temperatures.

Differential thermal analysis is a laboratory method which is especially useful in studying high-temperature reactions in clays because the deflections in the curve graphically represent changes in phase within the material. This method analyzes substances under nonequilibrium conditions, and corroborating x-ray data must therefore be obtained under similar conditions, by air-quenching samples at the desired temperatures. If samples are "soaked" thermally, equilibrium conditions are approached; differences are then observed in phases and temperatures of formation between soaked samples and air-quenched samples.

All thermal curves for kaolinite and halloysite show two significant features below 1000° C.: (1) a prominent endothermic reaction between 500° and 600° C. which can be correlated with the absorption of energy that results from loss of hydroxyl groups; (2) an intense exothermic

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reaction just below  $1000^{\circ}\text{C}$ . which has been attributed to the crystallization of either mullite or what has been called  $\gamma\text{-Al}_2\text{O}_3$ .

The firing product at temperatures between the two prominent thermal peaks is referred to as "metakaolin" and "pro-mullite." The theory (Insley and Ewell (1)) which correlates the exothermic peak with  $\gamma\text{-Al}_2\text{O}_3$  implies that an intimate mixture of amorphous alumina and amorphous silica is formed after the lattice is dehydrated, and that  $\gamma\text{-Al}_2\text{O}_3$  is an intermediate step in the formation of mullite.

The mullite hypothesis (Comeforo, Fischer, and Bradley (2)) postulates that when hydroxyl groups are expelled, the noncrystalline compound which is formed possesses some residual structure. On further heating, the noncrystalline compound collapses into crystalline mullite nuclei which are then free to grow normally.  $\gamma\text{-Al}_2\text{O}_3$  is considered incidental and not a step in the major reactions of the firing process.

#### SELECTION OF KAOLINITE SAMPLES

Kaolinite is known to vary in degree of crystallinity because of random displacement of structural layers by multiples of  $b_0/3$ ; transitional types range from well crystallized to poorly crystallized. The diffraction diagrams of poorly crystallized kaolinite (similar to Brindley's (3) fireclay material) contain fewer reflections than those for well crystallized material. The degree of crystallinity should be known before kaolinite samples are fired, because the results of firing may vary at certain temperatures according to variation in crystallinity.

Different sequences of phase changes during firing may also develop depending upon the type of impurity in the clay. For this reason, the impurities associated with the clay, both kind and quantity, are even more important than crystallinity. Coarse particles can be removed by fractionation, and the finer impurities can usually be detected by  $x$ -ray methods. Cations which cause fluxing may be detected by chemical analysis.

A micaceous impurity, apparently intergrown with kaolinite, is often observed; it is probably related genetically or diagenetically to the kaolinite. (Kaolinites which contain mica that can be observed in diffraction patterns are not discussed here.) Particularly important are poorly crystallized kaolinites in which the micaceous material is not detected in powder diffraction patterns but in which it appears as an anhydride after the kaolinite is dehydrated.

Powder diffraction patterns of unfired kaolinite and kaolinite fired to  $800^{\circ}\text{C}$ . are shown in Fig. 1. The unfired clay shows no effects of mica, but at  $800^{\circ}\text{C}$ . the anhydride shows clearly. A few anatase lines are also present.



FIG. 1. X-ray powder diffraction photographs of mica-bearing, poorly crystallized kaolinite. A. Unfired clay. B. Fired to 800° C.

Undetected mica and other impurities probably caused some of the differences previously reported in the phase changes during firing and shown in thermal curves for kaolinite. Thus it is important to determine not only the degree of crystallinity, but also the kind and amount of impurities, detrital, intergrown, and chemical.

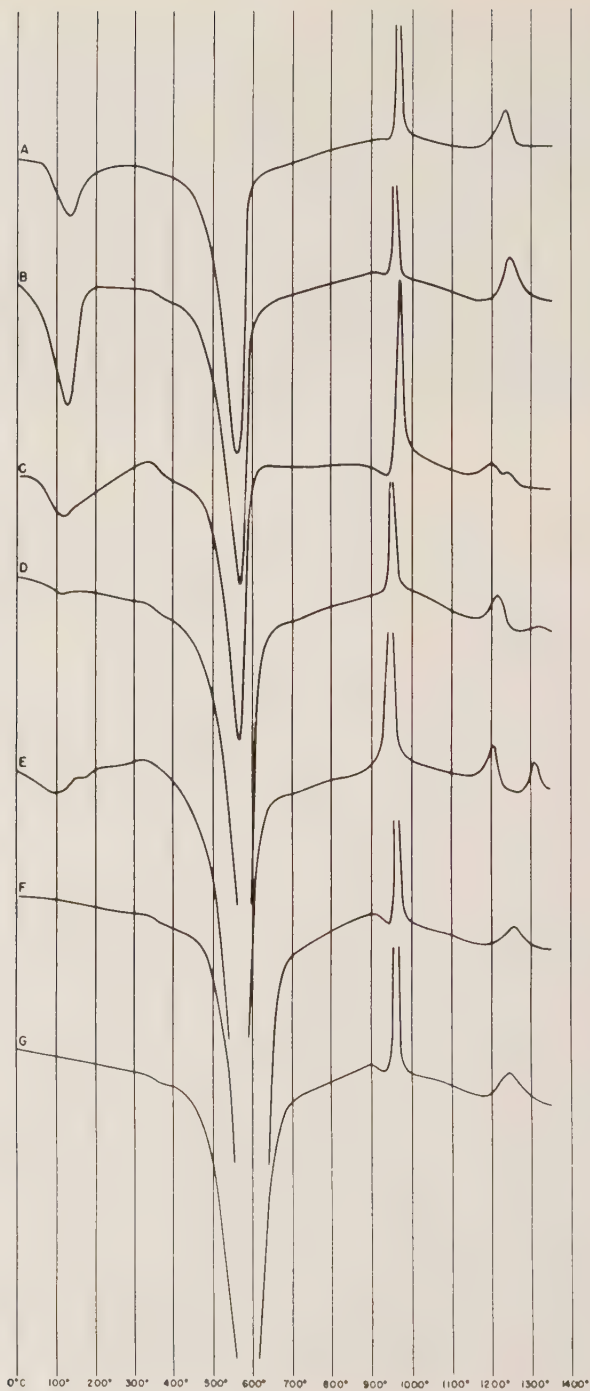
This study investigates the phase changes of mica-free, well crystallized kaolinite and mica-free, poorly crystallized kaolinite, mica-bearing, poorly crystallized kaolinite, and halloysite. No studies were made with kaolinite whose diffraction patterns showed visible mica, although such mica would be expected to cause additional effects on the thermal curve and firing sequence.

#### EXPERIMENTAL

Samples studied were mica-free, poorly crystallized kaolinite from Mt. Savage, Maryland; mica-bearing, poorly crystallized kaolinite from Anna, Illinois, and Sayreville, New Jersey; mica-free, well crystallized kaolinite from Dry Branch, Georgia, and Lewistown, Montana; halloysite ( $4\text{H}_2\text{O}$ ) from Bedford, Indiana, and halloysite ( $2\text{H}_2\text{O}$ ) from Mica, Washington.

The degree of crystallinity of the kaolinites and the physical impurities in all samples were determined by x-ray diffraction. The kaolinites contained minor anatase as an impurity, and the Illinois and Maryland clays contained a small amount of fine-grained quartz. The Maryland clay contained a small amount of an unknown impurity. The halloysites were free from impurity.

Thermal curves for all samples were run to 1350° C. and rerun for reproducibility checks. Samples were heated in the thermal furnace at a





rate of about  $10^{\circ}$  C. a minute to significant portions of the thermal curve below  $1000^{\circ}$  C. and also to  $1000^{\circ}$ ,  $1100^{\circ}$ ,  $1150^{\circ}$ ,  $1200^{\circ}$ ,  $1250^{\circ}$ ,  $1300^{\circ}$ , and  $1350^{\circ}$  C. The air-quenched samples were then examined by x-ray powder diffraction analysis.

Thermal curves are shown in Fig. 2. The phases for fired samples are listed in Table 1. Thermal curves and phases for both examples of each of the three pairs of clay types are identical except for the effects of quartz in the Illinois clay.

### THE THERMAL CURVES

There is no reason to expect that  $b_0/3$  shifts in the lattice structure of kaolinite should be reflected in the shape of the thermal curve. The principal features cited by Grim (4) as thermal differences between well and poorly crystallized kaolinite at temperatures below  $1000^{\circ}$  C. are as follows: (1) The poorly crystallized kaolinites have broad low-intensity initial endothermic reactions whereas the well crystallized kaolinites do not; (2) the intensity of the large endothermic reaction at about  $600^{\circ}$  C. and the final exothermic reaction are much greater for the well crystallized varieties; (3) the final exothermic reaction for well crystallized kaolinite is immediately preceded by a slight endothermic reaction whereas no such reaction is shown for poorly crystallized kaolinite. Other features may also be observed: the  $600^{\circ}$  C. endothermic peak is higher in temperature and occurs over a greater temperature range for well crystallized varieties; the exothermic slope of the curve between  $600^{\circ}$  and  $900^{\circ}$  C. is greater for well crystallized kaolinite, and the final exothermic peak is higher in temperature.

The smaller particle size of poorly crystallized kaolinite readily accounts for most of the differences except (3) above and the slope between  $600^{\circ}$  and  $900^{\circ}$  C. The important difference is the presence or absence of the small endothermic reaction which precedes the final exothermic peak. In Fig. 2 the peak is present in both mica-free, well crystallized kaolinite and mica-free poorly crystallized kaolinite and therefore cannot be related to the structure of the clay. The peak is absent in mica-bearing kaolinites because mica suppresses the peak. Absence of this peak indicates that the material is not pure.

Thus it appears that previous interpretations have been made on



FIG. 2. Differential thermal curves of kaolinite and halloysite. A. Halloysite ( $2\text{H}_2\text{O}$ ), Mica, Washington. B. Halloysite ( $4\text{H}_2\text{O}$ ), Bedford, Indiana. C. Mica-free poorly crystallized kaolinite, Mt. Savage, Maryland. D. Mica-bearing poorly crystallized kaolinite, Sayreville, New Jersey. E. Mica-bearing poorly crystallized kaolinite, Anna, Illinois. F. Mica-free well crystallized kaolinite, Lewistown, Montana. G. Mica-free well crystallized kaolinite, Dry Branch, Georgia.

TABLE 1. PHASES AND OBSERVED RELATIVE INTENSITIES FOR KAOLINITE AND HALLOYSITE FIRED UNDER NON-EQUILIBRIUM CONDITIONS

Temperature	800° C.	Exothermic peak ca. 950° C.	1000° C.	1100° C.	1150° C.	1200° C.	1250° C.	1300° C.	1350° C.
Mica-free well-crystallized kaolinite	metakaolin	amor. silica $\gamma$ -Al <sub>2</sub> O <sub>3</sub> mullite* <i>m w</i>	amor. silica $\gamma$ -Al <sub>2</sub> O <sub>3</sub> mullite <i>m w</i>	amor. silica $\gamma$ -Al <sub>2</sub> O <sub>3</sub> mullite <i>m w</i>	amor. silica $\gamma$ -Al <sub>2</sub> O <sub>3</sub> mullite <i>m w</i>	amor. silica $\gamma$ -Al <sub>2</sub> O <sub>3</sub> mullite <i>m</i>	? $\gamma$ -Al <sub>2</sub> O <sub>3</sub> mullite <i>w ms</i>	mullite <i>s</i>	mullite <i>s</i>
Mica-free poorly-crystallized kaolinite	metakaolin	amor. silica $\gamma$ -Al <sub>2</sub> O <sub>3</sub> mullite <i>m w</i>	amor. silica $\gamma$ -Al <sub>2</sub> O <sub>3</sub> mullite <i>m w</i>	amor. silica $\gamma$ -Al <sub>2</sub> O <sub>3</sub> mullite <i>m</i>	amor. silica $\gamma$ -Al <sub>2</sub> O <sub>3</sub> mullite <i>m</i>	mullite <i>ms</i>	mullite cristobalite <i>ms</i>	mullite cristobalite <i>ms</i>	mullite cristobalite <i>s</i>
Mica-bearing poorly-crystallized kaolinite	metakaolin $\alpha$ -SiO <sub>2</sub> † anhydride <i>m s</i>	$\alpha$ -SiO <sub>2</sub> anhydride $\gamma$ -Al <sub>2</sub> O <sub>3</sub> <i>s m</i>	$\alpha$ -SiO <sub>2</sub> $\gamma$ -Al <sub>2</sub> O <sub>3</sub> <i>s m</i>	$\alpha$ -SiO <sub>2</sub> $\gamma$ -Al <sub>2</sub> O <sub>3</sub> <i>s m</i>	$\alpha$ -SiO <sub>2</sub> $\gamma$ -Al <sub>2</sub> O <sub>3</sub> mullite <i>s w</i>	$\alpha$ -SiO <sub>2</sub> mullite <i>ms</i>	$\alpha$ -SiO <sub>2</sub> mullite cristobalite <i>w w</i>	mullite cristobalite <i>ms</i>	mullite cristobalite <i>s</i>
Halloysite	metakaolin	amor. silica $\gamma$ -Al <sub>2</sub> O <sub>3</sub> <i>m</i>	amor. silica $\gamma$ -Al <sub>2</sub> O <sub>3</sub> <i>m</i>	amor. silica $\gamma$ -Al <sub>2</sub> O <sub>3</sub> <i>m</i>	amor. silica $\gamma$ -Al <sub>2</sub> O <sub>3</sub> <i>m</i>	amor. silica $\gamma$ -Al <sub>2</sub> O <sub>3</sub> mullite <i>m w</i>	$\gamma$ -Al <sub>2</sub> O <sub>3</sub> mullite <i>w w</i>	mullite <i>s</i>	mullite <i>w</i>

\* Intensities of diffraction lines are not proportional to amounts.

† Not present in Sayreville, New Jersey, kaolinite.

impure materials, and there is no significant difference in the thermal curves for poorly crystallized and well crystallized kaolinite except for the effects caused by particle size.

Additional evidence for the presence of mica is indicated by the small endothermic reaction at about 700° C. which is present only in mica-bearing, poorly crystallized kaolinite. The reaction is caused by the dehydration of the micaceous intergrowth, and the peak indicates the presence of the anhydride. The peak is 150° C. higher than that observed for illite (5); dehydration at higher temperatures is evidence that there are muscovite-crystallization clay-micas intimately associated with kaolinite whose physical properties and composition differ from those of the original illite materials.

Above 1000° C., well crystallized kaolinite and halloysite show one exothermic peak at about 1250° C.; the poorly crystallized kaolinites show two distinct peaks. Mica-bearing varieties have peaks at about 1200° and 1300° C., and the mica-free clay has peak temperatures at about 1200° and 1240° C. The significance of the peaks is discussed below.

Characteristics of the halloysite thermal curve are: (1) there is a large initial endothermic peak at about 100° C.; (2) the large endothermic peak near 600° C. occurs at approximately the same temperature as the similar peak for poorly crystallized kaolinite but is reduced in intensity; (3) the peak for halloysite returns to base line faster than it departs, whereas well crystallized kaolinites show about equal rates of speed; (4) the exothermic slope above 600° C. is similar to the slope for mica-bearing, poorly crystallized kaolinite; (5) the small endothermic reaction at about 950° C. is less intense than it is for well crystallized kaolinite; (6) the exothermic reaction below 1000° C. is less intense than it is for well crystallized kaolinite; (7) the curve above 1000° C. is similar to that of well crystallized kaolinite, and the peak at 1250° C. is greater in intensity.

#### SEQUENCES OF PHASE CHANGE

For kaolinite and halloysite, the development of high-temperature phases depends on the type and amount of impurities as well as the crystallinity and morphology of the clay.

(1) *Mica-free kaolinite*.—Diffraction patterns of well and poorly crystallized kaolinites are identical up to 1000° C., which indicates that neither structure nor impurities exerts influence below this temperature. Diffraction patterns made after loss of hydroxyl groups are characterized by a broad diffuse asymmetrical reflection at about 4.3 a.u. as well as at least two weaker bands at higher diffraction angles which could not be measured. These reflections are attributed to the noncrystalline com-

pound of metakaolin (2). As the samples were mica-free, there was no anhydride structure. The 960° C. exothermic peak coincides with the formation of mullite,  $\gamma\text{-Al}_2\text{O}_3$  and amorphous silica; the amount of  $\gamma\text{-Al}_2\text{O}_3$  formed is less than that of mullite. The amount of mullite observed in diffraction patterns depends upon the crystallinity of the clay; diffraction intensities are greater in well crystallized varieties. The low-temperature mullite nuclei are greatly elongated along the *c*-axis and grow slowly, mostly in lateral directions, with increase of temperature (2). The 960° C. exothermic peak also coincides with a decrease in intensity of the 4.3 a.u. band.

Above 1000° C. the temperatures at which phases form apparently depend on impurities. The 1200° C. exothermic peak for poorly crystallized kaolinite and the 1250° C. peak for well crystallized kaolinite are correlated with a second generation of mullite from  $\gamma\text{-Al}_2\text{O}_3$  and amorphous silica.  $\gamma\text{-Al}_2\text{O}_3$  diffraction lines and the reduced 4.3 a.u. band retain their intensities until the start of the thermal peak; their disappearance at the peak is accompanied by a sharp increase in intensity of mullite diffraction lines. The higher-temperature mullite crystals do not exhibit the pronounced elongation of the low-temperature nuclei.

After mullite has formed from  $\gamma\text{-Al}_2\text{O}_3$  and amorphous silica, the remaining silica crystallizes to form cristobalite. The process is indicated by the small exothermic peak at temperatures higher than the upper mullite thermal peak. For well crystallized varieties cristobalite does not form within the temperature range studied and therefore no exothermic peak was observed. The reaction evidently occurs at temperatures above 1350° C. (6). On the other hand, poorly crystallized kaolinite formed cristobalite at about 1240° C. and the thermal peak is therefore present.

(2) *Mica-bearing kaolinite*.—The sequence of high-temperature phases is greatly affected by mica. Diffraction patterns made after loss of hydroxyl groups indicate the presence of a crystalline three-layer anhydride. No micaceous impurity could be detected in powder diffraction diagrams or by petrographic analysis of unfired samples, but diffraction effects from well-oriented aggregates revealed a small amount of micaceous material. For powder diagrams, the anhydride is apparent only after the kaolinite structure is destroyed by firing. Analytical data indicate 0.53 and 0.60 per cent  $\text{K}_2\text{O}$  for the Illinois and New Jersey clays respectively, not unreasonable figures for small amounts of mica.

As powder x-ray and petrographic methods may not detect small amounts of mica, they may not necessarily be accurate methods for establishing purity of kaolinite.

The anhydride structure persists to the 950° C. exothermic peak, and only  $\gamma\text{-Al}_2\text{O}_3$  and amorphous silica is observed in diffraction patterns at this temperature. The destruction temperature is about 100° C. higher



than those observed by Grim and Bradley (5) for the original illite materials and is probably related to the different mode of origin of the two micaceous materials. No mullite phase was observed at the 950° C. peak.

The 1200° C. thermal peak is related to second generation mullite formation from  $\gamma\text{-Al}_2\text{O}_3$  and amorphous silica. The typical sequence again occurs, mullite diffraction lines appear at the peak where  $\gamma\text{-Al}_2\text{O}_3$  lines and 4.3 a.u. band disappear. The excess silica, converted to cristobalite at about 1300° C., is represented by a small thermal peak. Cristobalite does not form until all  $\gamma\text{-Al}_2\text{O}_3$  has been used up in mullite formation. The mullite peak at 1200° C. is about 50° C. lower in impure kaolinites. There is evidently a relationship between the peak temperatures for mullite and cristobalite. Whenever the mullite peak is formed at about 1200° C. the cristobalite peak occurs within the temperature range studied (1240°–1320° C.). But for pure clays the mullite peak occurs at about 1250° C. and no cristobalite peak is present up to 1350° C.

(3) *Halloysite*.—The firing sequence for halloysite is identical with that of pure kaolinite up to the 960° C. exothermic peak. For halloysite,  $\gamma\text{-Al}_2\text{O}_3$  and amorphous silica are observed in diffraction patterns at this temperature, similar to mica-bearing, poorly crystallized kaolinite.  $\gamma\text{-Al}_2\text{O}_3$  and the 4.3 a.u. band disappear as mullite is formed at the 1250° C. exothermic peak. The larger size of the upper halloysite mullite peak as compared to that for well crystallized kaolinite is evidently caused by the crystallization of a greater amount of mullite at 1250° C. Thus the observed firing sequence for halloysite and poorly crystallized micaceous kaolinite is similar, the second mullite peak for halloysite being about 50° C. higher. As with well crystallized kaolinite, no cristobalite is formed up to 1350° C., and the excess silica remains amorphous.

#### THE DEHYDRATED STATE

There are two possibilities for the structure of kaolin-type minerals after loss of hydroxyl groups: (1) an intimate mixture of amorphous alumina and amorphous silica; (2) an amorphous compound of alumina and silica (metakaolin). The evidence of the 4.3 a.u. and related bands in diffraction patterns implies that a noncrystalline compound is formed because these bands are more prominent than those observed from truly amorphous solids (2). It has been demonstrated by Brindley (7) that single crystal photographs of dehydrated nacrite show a coordinated structure and not an amorphous mixture. The metakaolin structure has been interpreted by Comeforo, Fischer and Bradley (2) as "coordinated strings of alumina octahedra, with only some buckling of the silica layer to provide the additional oxygen necessary to complete the arrangement."

The diffraction effects of such a structure would be identical with those to be expected from amorphous silica (8). In metakaolin the silica is confined to the structure, but at temperatures following the major exothermic peak, the compound has been destroyed and amorphous silica exists with  $\gamma\text{-Al}_2\text{O}_3$ . The decreased intensity 4.3 a.u. band here indicates that amorphous silica is present.

#### THE MAJOR EXOTHERMIC PEAK

The phases at the major exothermic peak for pure kaolinite show that although  $\gamma\text{-Al}_2\text{O}_3$  and amorphous silica are always present, mullite is the dominant phase. Experiments which have shown only  $\gamma\text{-Al}_2\text{O}_3$  at the prominent peak have evidently used mica-bearing kaolinite. As shown in Table 1, only  $\gamma\text{-Al}_2\text{O}_3$  is present in diffraction patterns at this temperature for mica-bearing varieties.

The formation of a hard dense phase such as mullite more readily explains the large evolution of energy than does a subcrystalline compound such as  $\gamma\text{-Al}_2\text{O}_3$ . The early mullite crystals are asymmetrical and strongly suggest that "long strings of alumina coordination octahedra are carried over directly from the one structure to the other" (2). As the earlier mullite formation is determined by the structure of the original kaolinite, any possibility of an intermediate phase such as  $\gamma\text{-Al}_2\text{O}_3$  is eliminated.

The formation of mullite from the collapse of the metakaolin structure cannot use up all alumina, as shown by the formation of  $\gamma\text{-Al}_2\text{O}_3$  at the peak. An excess of silica is also indicated by the decreased intensity 4.3 a.u. band. Thus the formation of mullite directly from the metakaolin phase leaves the remaining silica and alumina as separate oxides in the form of amorphous silica and  $\gamma\text{-Al}_2\text{O}_3$ . The phases at the exothermic peak for clean kaolinites should always show the three phases: mullite,  $\gamma\text{-Al}_2\text{O}_3$ , and amorphous silica. There is no amorphous alumina phase which forms  $\gamma\text{-Al}_2\text{O}_3$ , so that the  $\gamma\text{-Al}_2\text{O}_3$  for the first formation of mullite is an accompanying phase and not an intermediate phase.

The growth of mullite nuclei is predisposed by the structure of the clay mineral fired. Two factors must be taken into consideration: (1) the degree of order in the stacking of the layers; (2) the morphology of the crystal lattice. The amount of mullite detected at the exothermic peak decreases as the structure becomes more random, and the thermal peak decreases in intensity. The  $b_0/3$  shifts in kaolinite evidently cause the development of smaller mullite nuclei with resultant decrease in thermal intensity.

For halloysite, the effect of morphology is pronounced. In the dehydrated kaolinite structure, coordinated alumina and silica layers are of much greater lateral extent than in the halloysite metakaolin phase. Breakdown of the kaolinite structure affords little opportunity for the

alumina from one sheet to combine with alumina from other sheets to form  $\gamma\text{-Al}_2\text{O}_3$  except at the edges of the layers. In halloysite, the smaller dimensions of the layers permit much greater interaction of alternate alumina layers resulting in a decreased particle size of the primary mullite nuclei. Mullite nucleation could hardly be expected to pass any grain boundary and thus might well consist of too few units to afford recognizable x-ray diffraction effects. The mullite nuclei are too small to be registered by x-ray diffraction, and  $\gamma\text{-Al}_2\text{O}_3$  is the only crystalline phase actually observed. This results in a decreased intensity of the thermal peak although the formation of mullite may well be the cause for the effect.

For mica-bearing kaolinite, the effect of the intergrown micaceous anhydride may also inhibit the size of early mullite crystals, so that only  $\gamma\text{-Al}_2\text{O}_3$  is revealed in diffraction patterns and in reduced intensity of the thermal peak.

Primary mullite forms directly from the metakaolin structure at the major exothermic peak and is observed in diffraction patterns of mica-free kaolinite but not in mica-bearing kaolinite or halloysite. Thus it appears that morphology and impurities determine whether or not mullite will be detected in diffraction patterns at the major exothermic peak.

Although the alumina phase at the major exothermic peak is called  $\gamma\text{-Al}_2\text{O}_3$  in this study, there are differences between it and  $\gamma\text{-Al}_2\text{O}_3$  formed in the firing of aluminum hydrates. The diffraction lines of alumina from kaolin-type minerals are symmetrical reflections, whereas those of aluminum compounds are asymmetrical. The kaolin-type alumina phase more nearly resembles a spinel structure than does pure  $\gamma\text{-Al}_2\text{O}_3$ . Furthermore, the size of the unit cell is slightly larger than that of pure  $\gamma\text{-Al}_2\text{O}_3$ .

#### SECONDARY MULLITE AND CRISTOBALITE

As has been shown, formation of primary mullite coincides with the formation of  $\gamma\text{-Al}_2\text{O}_3$  and amorphous silica. The latter phases persist until the second exothermic reaction, at temperatures between  $1200^\circ$  and  $1250^\circ\text{C}$ ., depending on the type of clay fired. The disappearance of the diffraction lines of the silica and alumina phases at this point and the accompanying increase in amount and sharpness of symmetrical mullite lines indicates that a second generation of mullite has been accompanied by growth of the primary mullite nuclei. The higher temperature mullite forms from  $\gamma\text{-Al}_2\text{O}_3$  and amorphous silica, and the alumina is now an intermediate phase in mullite formation. In halloysite and mica-bearing kaolinite, the early crystallites do not develop until the reaction between  $\gamma\text{-Al}_2\text{O}_3$  and amorphous silica takes place. The  $1250^\circ\text{C}$ . mullite thermal

peak for halloysite is more intense than the similar peak for well crystallized kaolinite, and the 1200° C. mullite peak for mica-bearing kaolinites is more intense than the similar peak for mica-free, poorly crystallized kaolinite. This indicates a sudden growth of primary crystallites as well as crystallization from  $\gamma$ -Al<sub>2</sub>O<sub>3</sub> and amorphous silica for those clays which show no mullite in diffraction diagrams at the major exothermic peak.

The amorphous silica which remains after formation of secondary mullite forms cristobalite at the exothermic peak above the second mullite peak. Within the temperature range used, cristobalite peaks were observed only in impure specimens. The upper mullite peak is 50° C. lower in the impure poorly crystallized kaolinites, and the cristobalite peak is well below 1350° C.

The presence or absence of mullite in diffraction patterns at the major exothermic peak is not related to formation of secondary mullite at 1200° or 1250° C. Primary mullite is observed only for mica-free kaolinites, but the poorly crystallized variety forms secondary mullite at 1200° C. and the well-crystallized clays at 1250° C.

The organization of the unfired clay does not explain the difference in temperature for the second mullite peak. The clay with the highest degree of order in the stacking of structural units (well-crystallized kaolinite) and that with the lowest degree of order (halloysite) both have upper mullite peaks at 1250° C.

The variation in temperature may therefore be caused by impurities. Physical impurities such as quartz will not cause any lowering in temperature of peaks. It is thus possible that potassium and other fluxing elements present in mica and other impurities may be the cause for the variation.

In general, presence of fluxing impurities lowers the temperature of formation of secondary mullite and cristobalite, and it is possible that increase in mica could cause further changes limited by the fluxing action of potassium.

When the secondary mullite peak occurs at about 1200° C. and the cristobalite peak below 1350° C. the inversion of small amounts of quartz to cristobalite is always accompanied by the crystallization of cristobalite from amorphous silica. If large amounts of quartz impurity are present (amount unknown), no inversion occurs within the temperature range studied, nor does crystallization from amorphous silica take place. The thermal curve then shows only one peak at about 1200° C.

For clays where the mullite peak occurs at about 1250° C., no cristobalite peak is formed regardless of how much quartz impurity is present.

Although the formation of cristobalite from the crystallization of amorphous silica and the inversion of quartz occur at the same tempera-



ture, the thermal effect observed is caused only by amorphous silica crystallization. The inversion from quartz is an endothermic reaction and is not apparent because of the greater amplitude of the crystallization effect.

#### EQUILIBRIUM FIRING CONDITIONS

The phase sequences reported above represent changes during firing under non-equilibrium conditions at a constant heating rate. If samples are soaked or the rate of heating is varied, phases occur at different temperatures from those under the experimental conditions reported here. Thus micaceous, poorly crystallized kaolinite, which shows only  $\gamma\text{-Al}_2\text{O}_3$  at  $1100^\circ\text{C}$ . when air-quenched, shows a complete mullite pattern with minor cristobalite after it soaks for 4 hours at  $1100^\circ\text{C}$ . Under this condition, mullite forms at temperatures  $100^\circ\text{C}$ . less than when air-quenched; and cristobalite forms at  $200^\circ\text{C}$ . less. Cristobalite, not observed in well crystallized, mica-free kaolinite at  $1350^\circ\text{C}$ . under non-equilibrium conditions, can be developed at lower temperatures by soaking. Prolonged firing, therefore, lowers the temperatures at which phases form and decreases the total temperature interval in which a complete firing sequence occurs. Only under non-equilibrium conditions can the phases be investigated without confusion.

The development of phases is directly dependent upon 4 factors: rate of heating, nature of equilibrium conditions, amount and type of impurity, and structure. The lack of agreement in the literature as to the sequence and interpretation of phase changes is due to the failure to control some or all of the four variables.

#### SUMMARY

Thermal curves for kaolinite and halloysite to  $1350^\circ\text{C}$ . are dissimilar enough for precise identification. The essential differences between the thermal curves of well and poorly crystallized kaolinites to  $1000^\circ\text{C}$ . are the effects principally of decreased particle size.

After dehydration, an amorphous compound of silica and alumina (metakaolin) forms, which persists to the major exothermic thermal peak, then collapses to form mullite nuclei,  $\gamma\text{-Al}_2\text{O}_3$ , and amorphous silica. The amount and size of the mullite nuclei depend upon the morphology, structure and purity of the fired material. Mullite formation causes the principal evolution of energy.  $\gamma\text{-Al}_2\text{O}_3$  is not an intermediate step in mullite formation at this temperature.

The development of phases at higher temperatures is controlled principally by fluxing impurities. Secondary mullite occurs at about  $1200^\circ\text{C}$ . for impure clays and at about  $1250^\circ\text{C}$ . in clean samples; it forms from  $\gamma\text{-Al}_2\text{O}_3$  and amorphous silica. At this point  $\gamma\text{-Al}_2\text{O}_3$  is an

intermediate step in mullite formation. The silica which remains forms cristobalite between 1240° and 1320° C. in impure clays and probably above 1350° C. in pure clays. Fluxing impurities tend to lower the temperature of thermal peaks for mullite and cristobalite.

Variation in amount and type of impurities, rate of heating, degree of equilibrium reached during firing, and structure of the clay all affect the sequence and temperature of phase formation. Failure to standardize firing conditions is a primary cause of lack of agreement in the literature as to the firing sequence of kaolinite and halloysite.

The phases observed in diffraction diagrams are as follows:

	Lattice dehydration	Major exothermic peak	Mullite thermal peak	Cristobalite thermal peak
Mica-free well crystallized kaolinite	metakaolin	mullite $\gg \gamma\text{-Al}_2\text{O}_3$	1250° C.	none present to 1350° C.
Mica-free poorly crystallized kaolinite	metakaolin	mullite $> \gamma\text{-Al}_2\text{O}_3$	1200° C.	1240° C.
Mica-bearing poorly crystallized kaolinite	metakaolin, mica-anhydride	$\gamma\text{-Al}_2\text{O}_3$ , mica-anhydride	1200° C.	1300° C.
Halloysite	metakaolin	$\gamma\text{-Al}_2\text{O}_3$	1250° C.	none present to 1350° C.

The effect of heat on kaolin-type materials may be expressed as follows:

Above 600° C.	$\text{Al}_2\text{O}_3 \cdot 2\text{SiO}_2 \cdot 2\text{H}_2\text{O} \rightarrow \text{Al}_2\text{O}_3 \cdot 2\text{SiO}_2 + 2\text{H}_2\text{O}$ kaolin metakaolin
950°–980° C.	$\text{Al}_2\text{O}_3 \cdot 2\text{SiO}_2 \rightarrow 3\text{Al}_2\text{O}_3 \cdot 2\text{SiO}_2 + \gamma\text{-Al}_2\text{O}_3 + \text{SiO}_2$ primary mullite gamma alumina amorphous silica
1200°–1250° C.	$\gamma\text{-Al}_2\text{O}_3 + \text{SiO}_2 \rightarrow 3\text{Al}_2\text{O}_3 \cdot 2\text{SiO}_2 + \text{SiO}_2$ secondary mullite amorphous silica
1240°–1350° C. (?)	$\text{SiO}_2 \rightarrow \text{SiO}_2$ cristobalite

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# WHEWELLITE FROM A SEPTARIAN LIMESTONE CONCRETION IN MARINE SHALE NEAR HAVRE, MONTANA\*

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## ABSTRACT

Whewellite, calcium oxalate monohydrate, has been found as a crystal perched on terminated, unetched, drusy calcite crystals in a septarian limestone concretion in the marine Bearpaw shale of Late Cretaceous age near Havre, Montana. Its occurrence resembles that in marl concretions in the Caucasus but differs from occurrences in other regions. The inorganic precipitation of whewellite probably occurred in an alkaline environment, and the oxalate was probably derived from nearby organic material through the action of ground water. Other geological occurrences of whewellite are reviewed and the conclusion expressed that hypotheses suggesting hydrothermal derivation of the oxalates or their natural precipitation in deep sea sediments need confirmation.

## INTRODUCTION

In June 1952 Robert B. McGraw, Jr., collected from a septarian limestone concretion near Havre, Montana, a vuggy specimen of yellow calcite containing a mineral since identified as whewellite, calcium oxalate monohydrate. This is the first North American locality reported for the mineral. In casual field examination, whewellite might easily be mistaken for calcite or barite; so a detailed account of its occurrence and identity is recorded here as an aid to future discoveries of the mineral in the same or similar environment. Dr. George Switzer of the U. S. National Museum provided for comparative study a specimen from Pchery, Schlan, Bohemia (Czechoslovakia), the occurrence of which is described by Slavik (1908). The specimen from the new locality described in this paper has been deposited in the U. S. National Museum as specimen No. 106555.

## OCCURRENCE

A single crystal of whewellite was found in a fossiliferous, septarian limestone concretion in the marine Bearpaw shale of Late Cretaceous age near the center of sec. 33, T. 32 N., R. 17 E., about 7 miles southeast of Havre, Montana, via the Havre-Clear Creek road, and about 300 feet southeast of a bridge that crosses Statten Coulee. Here several limestone concretions 1 to 4 feet in diameter are exposed in the shale. The enclosing shale is dark gray, for the most part fissile, and locally bentonitic and gypsiferous. Some of the concretions are fossiliferous and many

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are septarian. The whewellite-bearing concretion contained invertebrate fossil shells.

Yellow to brownish-yellow calcite in irregular and interconnecting veins characterizes the marine septarian concretions of this region. Although minerals other than calcite are extremely rare in the concretions, barite, gypsum, and quartz have been collected from several. The general mineralogy and chemical composition of these concretions will be reported at some future time to augment the information already published by Rubey and Bass (1925), Cobban (1951), and others.

The concretion containing the whewellite had the form of an oblate spheroid with a diameter of about 2 feet and a thickness of 1 foot. The exterior surface is dark grayish brown and freshly broken surfaces are olive gray. The septarian vein development ranges in width from about 3 inches down to barely perceptible fractures lined with calcite. In contact with the rock of the concretion and separating it from the coarse calcite of the septarian veins is a thin layer of fibrous calcite ( $\omega = 1.659$ ), rarely more than 2 mm. thick, that fluoresces bright yellow in a short-

TABLE 1. PHYSICAL PROPERTIES OF WHEWELLITE

	Havre, Mont.	Burgk, Saxony	Maikop, Caucasus
$\alpha$	1.491	1.4909	1.49
$\beta$	1.555	1.5554	1.55
$\gamma$	1.654	1.6502	1.65
2V (+)	82°	83°55'	82.5°
Orientation	—	X=b Z $\wedge$ c = +30°	—
Gr.	2.21 $\pm$ 0.01	2.226	2.22
Hardness	3	—	3.5

wave ultraviolet light. The coarse, nonfluorescent calcite ( $\omega = 1.662$ ) crystals are yellow to pale yellow, attain a maximum diameter of 8 mm., and have rhombohedral terminations that project into the vuggy parts of the veins. Locally the veins of the coarse calcite transect cracks filled with the fibrous fluorescent calcite. Gypsum sparsely encrusts the coarse calcite crystals in some vugs and also has formed along cracks that transect both calcite layers. The whewellite crystal is perched on coarse calcite crystals that have rhombohedral terminations, are pale yellow, and show no evidence of etching. The surface of the whewellite is coated by a thin pink-colored incrustation of very fine grained aragonite and pink calcite that conceals the transparent interior of the mineral.

TABLE 2. MEASURED  $d$ -SPACINGS OF  $x$ -RAY POWDER PATTERNS OF WHEWELLITE FROM MONTANA AND SCHLAN, BOHEMIA (CZECHOSLOVAKIA)

Montana (USGS film No. 3960)		Schlan, Bohemia <sup>2</sup> (USGS film No. 3963)	
Intensity <sup>1</sup>	$d(\text{\AA})$	Intensity <sup>1</sup>	$d(\text{\AA})$
100	5.95	100	5.94
5	5.81	10	5.80
5	4.53	5	4.52
5	3.773	5	3.779
90	3.652	90	3.650
1	3.414	1	3.402
1	3.321	—	—
5	3.105	1	3.106
1	3.002	—	(F)
50	2.971	60	2.966
10	2.906	10	2.909
10	2.842	10	2.842
1	2.639	—	(F)
20	2.497	40	2.494
1	2.455	—	—
1	2.422	—	—
1	2.388	—	—
80	2.357	80	2.354
20	2.262	30	2.263
5	2.213	10	2.212
1	2.135	1	2.129
10	2.076	30	2.079
5	1.978	10	1.978
5	1.955	10	1.954
5	1.930	10	1.930
5	1.891	10	1.890
1	1.859	—	—
1	1.848	5	1.850
1	1.826	—	—
1	1.818	10	1.818
1	1.794	10	1.792
1	1.737	10	1.737
1	1.693	1	1.696
1	1.644	1	1.638
1	1.588	1	1.589
1	1.552	1	1.556
—	—	1	1.548
1	1.528	1	1.528
1	1.505	1	1.504
1	1.480	1	1.482
1	1.458	—	—

F, faint lines; not measured.

<sup>1</sup> Estimated.<sup>2</sup> U. S. National Museum specimen No. 86913.

## DESCRIPTIVE MINERALOGY

The whewellite crystal, as originally found in the concretion, had a prismatic habit and dimensions of 25 by 8 by 3 mm. Identification as whewellite was made from its optical data and  $x$ -ray powder-diffraction pattern. Its physical properties are compared in Table 1 with those reported by Jezek (1911, p. 18) for whewellite from Burgk, Saxony, and by Vassoyevicz and Razumovsky (1928) for whewellite from the Maikop region, northern Caucasus. A summary of properties for whewellite is given by Wherry (1922).

The hardness of whewellite from Montana is about 3 and its specific gravity, determined with a Berman balance, is 2.21. Morphology of the mineral is well described by numerous authors who had many crystals to work with.

$X$ -ray power-diffraction photographs of whewellite from Montana and from Pchery, Schlan, Bohemia, are almost identical; and since some of the previously published powder data for the monohydrate are incomplete or inaccurate, the  $d$ -spacings and intensities for both specimens are given in Table 2.

Honnegger (1952), for example, has tabulated powder data for whewellite from four different sources. Supposedly, only the stronger  $\alpha$ -reflections are included; however, the medium strong line with a  $d$ -spacing of 4.03–4.05 Å is almost certainly a  $\beta$ -reflection of the strong line with a  $d$ -spacing of 3.64 Å. This line was not observed on the photographs taken for this paper nor in several patterns which are included in the *A.S.T.M.* files. The  $d$ -spacings published in this paper were corrected for film shrinkage by the back reflection method and are given for many weaker lines which have not been previously listed for whewellite.

## CHEMICAL DATA

Whewellite, fluorescent and nonfluorescent calcite, and the pink coating on whewellite were submitted for spectrographic examination and the results are shown in Table 3. The whewellite is exceptionally pure. The calcite is ferroan, and the fluorescent variety has the more strontium, barium, and magnesium. No explanation for the pink color of the coating on the whewellite can be offered other than organic dye.

## GEOLOGICAL ENVIRONMENT

Whewellite and other hydrated oxalates have been reported from a wide variety of organic and inorganic environments. Oxalates are widely distributed in living plant tissues and have been identified in sedimentary beds containing lignite or other organic substances. Both the monohydrate and the dihydrate (weddelite) are associated with a great variety

TABLE 3. QUALITATIVE SPECTROGRAPHIC ANALYSES OF WHEWELLITE AND OTHER MINERALS FROM A LIMESTONE CONCRETION NEAR HAVRE, MONT.  
(R. S. Harner, *analyst*)

	Mn	Fe	Mg	Sr	Ba
1. Whewellite (clear, colorless)	—	0.00X	0.00X	0.00X	0.00X
2. Calcite (fluorescent variety)	0.0X	X	0.X	0.X	0.0X
3. Calcite (nonfluorescent variety)	0.X	X	0.0X	0.00X	0.00X
4. Pink coating on whewellite	0.00X	0.0X	0.X	0.0X	0.00X

Not found in samples: Cu, Ag, Au, Hg, Ru, Rh, Pd, Os, Ir, Pt, Mo, W, Re, Ge, Sn, Pb, As, Sb, Bi, Se, Te, Zn, Cd, Tl, In, Co, Ni, Al, Ga, Cr, V, Sc, Y, La, Ce, Ti, Zr, Hf, Th, Nb, Ta, U, Be.

of urinary calculi in mammals (see Prien and Frondel, 1947; Milton and Axelrod, 1951).

The hydrated calcium oxalates can be prepared by simple reaction between calcium salts and oxalates in water solution. Bramlette (Bradley and Bramlette, 1942, p. 29), for example, calls attention to the formation of hydrated calcium oxalate during mechanical analysis experiments where sodium oxalate was added to washed muds obtained from the North Atlantic floor. He verified their presence in the pipette samples but their absence in the untreated sediments. Crystals of calcium oxalate dihydrate (weddellite) were identified by Bannister (Earland, et al., 1936) in bottom sediments collected during the cruise of the *Scolia* in 1902 to 1904 in the Weddell Sea, Antarctica, at seven stations where depths ranged from 4,434 to 5,008 meters. Gypsum crystals were also identified in sediments collected on the same cruise at five stations; and earlandite, a hydrated calcium citrate, was found as warty nodules in sediments collected at one station. The natural occurrence of weddellite in bottom sediments of the deep Weddell Sea is accepted by Earland and Bannister, but perhaps some reasonable doubt may be raised in view of the description of the sample storage. Earland (Earland, et al., 1936, p. 6) states: "Most of the deposits received by me were in their original containers. . . . The containers were principally bottles and jars in which provisions had been preserved, many still bearing the original labels. Their condition varied, some being in perfect preservation after thirty years in store, while others, owing to defective corks, had dried up." The nature of the food originally stored in the containers is not mentioned.

Many excellent crystals of whewellite have been reported as attached to walls of vugs in a great number of carbonate-sulfide veins of mesothermal association in central Europe (Ungemach, 1909; Jezek, 1911; and others). In these deposits the mineral association includes carbon-



ates (calcite, siderite, ankerite, and dolomite), barite, and sulfides (sphalerite, pyrite, pyrrhotite, marcasite, chalcopyrite, tetrahedrite, millerite, and linnaeite). Ungemach (1909) accepts this association as evidence in support of the hydrothermal rather than supergene derivation of the oxalate, although the occurrences are not deep-seated and the veins traverse rocks that are locally lignitic or graphitic.

The occurrence of whewellite in calcite veins up to 2 cm. thick at a single locality in Devonian argillite in southern Timan is reported by Kalyuzhnyi (1948) who ascribes the origin of the oxalate to incomplete oxidation of organic material contained in the argillite itself.

A geological occurrence closely resembling that near Havre, Montana, is described by Vassoyevicz and Razumovsky (1928) in the Maikop district (northern Caucasus). Tabular, well-developed, wine-yellow crystals of whewellite occur in compact marl concretions that are abundant in the upper part of an Eocene foraminiferal claystone that is somewhat calcareous and bituminous. The origin of the oxalate is ascribed to oxidation of organic matter in the rock.

#### CONCLUSIONS

Precipitation of whewellite in a limestone concretion near Havre, Montana, took place well after the development of the septarian calcite veins. The unetched faces of the contiguous calcite crystals support the existence of an alkaline environment for the formation of the whewellite here.

Although hydrous oxalates are widely distributed in nature and have formed by organic as well as inorganic processes, hypotheses suggesting hydrothermal derivation of the oxalates or their natural precipitation in a deep sea environment to explain some inorganic occurrences may need further confirmation. Occurrences in consolidated sedimentary rocks are best explained by deposition from circulating ground water that has acquired oxalate from nearby organic material in the sediments.

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# THE PREPARATION OF SPECIMENS FOR THE FOCUSING-TYPE X-RAY SPECTROMETER

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## ABSTRACT

Following a discussion of the requirements for mounts that will yield high precision for  $x$ -ray diffraction spectra obtained with the focusing-type spectrometer, a method is described for preparing specimens pressed into metal holders, and details given concerning a suitable press with a rectangular mold. A finely powdered material is mixed with a small amount of Lucite solution, and after drying and crushing it is heated and pressed so as to obtain a flat surface in proper registry with the metal mount. Samples prepared in this way will permit full advantage to be taken of the remarkable precision of which modern spectrometers are capable.

## INTRODUCTION

The object of this paper is to describe a device for the rapid preparation of mounted specimens so as to obtain high precision with the focusing  $x$ -ray spectrometer. As is now well known, such spectrometers operate by means of a broad beam of  $x$ -rays emanating from a narrow source and impinging on the flat, or slightly curved, surface of the specimen. The geometrical arrangement of the set-up is such that the  $x$ -rays diffracted from the various planes in the crystalline particles are brought to a sharp focus at a certain distance from the specimen, where the lines corresponding to the various crystallographic planes are registered on a photographic film, or are allowed to operate a Geiger-Müller Counter, the impulses from which after suitable amplification produce a series of peaks on a chart recorder.

Specimens are commonly prepared by (1) mixing a small amount of the powdered material with a diluted lacquer, e.g., a 10 per cent by volume mixture of Duco cement in ethyl acetate or acetone and spreading the material so that it covers a suitable rectangular area on a glass microscope slide; (2) by dusting some of the powder in a rectangular area lightly coated with vaseline; (3) pressing the material with a spatula into a rectangular well on a glass or metal slide and smoothing the top surface to a plane with a suitable implement such as the edge of a glass slide; or (4) mixing the material with a "plastic" molding powder and forming it into a block by means of heat and pressure. Specimens prepared by any of these methods yield results that are fairly satisfactory for the identification of materials by the powder diffraction method, but, as shown below, the usual methods of preparation may introduce errors of as much as several hundredths of a degree in the Bragg angles and may thus reduce by a considerable amount the precision that may be

obtained with modern spectrometers. Not only will the possible error in the Bragg angle be diminished, but the better-prepared sample produces better resolution and more consistent and reliable intensities, these two considerations being of equal or greater importance than error in the Bragg angle. Moreover, a reproducible procedure for sample preparation is important in studying a suite of minerals. Otherwise, the investigator has no assurance that some variations in the patterns are not a result of hit-or-miss sample preparation.

We shall first consider briefly the principal requirements for satisfactory specimens and then proceed to describe the recommended method and device for preparation.

### REQUIREMENTS

For best results, it is imperative that the specimen have a flat surface and that this flat surface be positioned to satisfy the focusing requirements of the spectrometer (the surface lies in the plane of the top of the sample holder). Ideally, the surface should be cylindrical and of such a curvature that the corresponding circular arc would pass through the source, the center of rotation, and the receiving slit (or the image on the photographic film), but it can be shown (Wilson, 1950) that for most purposes a flat specimen is quite satisfactory, the displacement of the peaks by the lack of curvature being less than the usual error of measurement. Furthermore, only for special uses is there any important deterioration in sharpness of focus due to lack of curvature in the specimen.

On the other hand, the position of the face of the specimen is highly important. If this face does not pass through the axis of rotation—or does not at least bear a fixed relation to this axis—then large errors may be introduced. As demonstrated by Wilson (1950) the displacement in the Bragg angle  $2\theta$ , if the front face of the specimen is at distance  $s$ , from the axis of rotation, is  $-(2s/R) \cos \theta$ ,  $R$  being the spectrometer radius. This relation was tested by making measurements of the diffraction angles for silicon by “shimming” the specimen with strips of brass. Up to about one millimeter in thickness the displacement was found to be linear with respect to the thickness of the shims. The displacements in terms of  $2\theta$  of the  $26.47^\circ \alpha$  peak (Cu radiation) and  $88.03^\circ \alpha_1$  peak were found to be respectively  $0.66^\circ$  and  $0.48^\circ$  per millimeter. From Wilson's relation,  $R$  being 17 cm., the corresponding theoretical values,  $0.65^\circ$  and  $0.48^\circ$ , are readily obtained. Other types of  $x$ -ray spectrometers in common use have smaller values of  $R$ —and correspondingly greater displacement of the diffraction angles. It is apparent that, although the modern  $x$ -ray spectrometers of the focusing-type are capable of high



degrees of accuracy, such accuracy will not be obtained unless careful attention is paid to the registry of the specimen surfaces. To put the requirements on a numerical basis, we may note that the Norelco high-angle spectrometer with chart recording has a reading error of  $\pm 0.005^\circ$ , and according to Donnay and Donnay (1952), an accuracy of  $\pm 0.01^\circ$  when properly prepared specimens are used. This means that a lack of registry of the specimen surface in the amount of as little as 10 to 20 microns can produce noticeable errors in the diffraction angles.

Another requirement is that the material in the specimen should be as dense as possible. The reasons for this are as follows: Upon striking the sample, the beam of  $x$ -rays penetrates the surface by an amount that depends on the absorption coefficient of the material. The *effective* surface is thus below the real surface, and, in accordance with the relation mentioned above, the diffraction angles will be lowered. Theoretical studies by Wilson (1950) lead to the result that the lowering of the position of the "center of gravity" of the diffraction maximum is  $\sin 2\theta/4\mu_e R$ , where  $\mu_e$  is the linear absorption coefficient for the sample as prepared and for the appropriate wave length. Although the displacement of the actual peak is difficult to calculate, it appears that this displacement is usually somewhat smaller than the displacement of the center of gravity but of the same order of magnitude. With the elements of medium or high atomic weight and with wave length higher than  $1.5 \text{ \AA}$ , the error is small. For example, with copper K-radiation ( $1.5418 \text{ \AA}$ ) and with silicon, for which at this wave length the pressed powder has a value of  $\mu_e$  somewhat less than 100, the displacement due to the finite penetration of the sample is scarcely more than  $0.01^\circ$ ; but with materials with low atomic weight and with  $x$ -rays of low wave lengths, the absorption coefficients are small, and the displacements become large. It may be noted that at the wave length of molybdenum  $K\alpha$ -radiation ( $0.7107 \text{ \AA}$ ) the absorption coefficients are roughly one-tenth the magnitude of those for the copper  $K\alpha$ -radiation; and the displacements, therefore, about ten times greater. Some measurements on silicon powder with molybdenum  $K\alpha$ -radiation gave angles ( $\alpha_i$ ) for the 111 and 331 spacings that are low by several hundredths of a degree ( $2\theta$ ), while with diamond powder and the same radiation the  $\alpha$  peak for the 311 spacing was  $0.4^\circ$  ( $2\theta$ ) too low. The theoretical relation gives even larger displacements. At any rate, this is an important source of error under certain conditions. Its bearing on sample preparation is that loosely packed powders or powders diluted with a large amount of low-absorption material will have a relatively small effective absorption coefficient and will show a correspondingly large displacement due to penetration. It is important therefore that the material be densely packed.

A desirable, though not necessary item is that the sample be rectangular in shape and of the proper size. Because the illuminated portion of the sample is a rectangle, shapes such as circles are wasteful of material. In this connection it may be interesting to note that one dimension of the rectangle is approximately the length of the divergence slit, the other dimension by the usual arrangement varies with the angle, being approximately  $2R\psi/(\sin 2\theta)$ , in which  $\psi$  is the divergence angle, and as before,  $R$  is the spectrometer radius. For  $\psi$  equal to  $1^\circ$  and  $R$  equal to 17 cm. the "length" of the illuminated area is 34 mm. at  $2\theta$  equal to  $10^\circ$  and somewhat less than 4 mm. at  $90^\circ$ . It should be noted that considerations of space usually limit the length to 20 or 25 mm.

Finally, there are the desirable features of ruggedness, permanence, and ease of handling.

#### METHOD OF SAMPLE PREPARATION

The methods mentioned above all have definite disadvantages. We experimented with all of these procedures and found them to be unsatisfactory for measurements of high precision. The mounts made with lacquer or Duco are not always flat enough for maximum definition. More important, the surfaces being raised above the fiduciary surface by one or two tenths of a millimeter, the angles are lowered by something like  $0.1^\circ$ . Vaseline mounts have these disadvantages, and, in addition, are not very durable. Specimens made by pressing powdered material into a well may, if great care is used, have surfaces that are flat and in proper registry, but—especially with fine powders—the "finite absorption" error may be unduly large on account of the lowered bulk density. Blocks made with molding powder required from 50 to 75 per cent by volume of the molding powder in order to obtain adequate strength. This great dilution of the material under investigation reduces the effective density and may lead to significant errors with materials of low absorption, especially when short wave lengths are employed.

The reason that a large proportion of molding powder is required to make a coherent cake is that even when the molding powder is finely ground it is difficult to obtain an intimate mixture of the two materials. This difficulty is circumvented by mixing the ground material with a thin solution of the molding powder, then drying, crushing, and pressing.

The sample preparation procedure outlined below produces effective samples conveniently and quickly. Samples prepared in this way give reliable results, meeting the accuracy requirements of our work.

The material, after being put through a 200-mesh screen, is given sufficient grinding with ethyl acetate in a corundum mortar so that the material is not noticeably gritty. (It is doubtful whether for the focusing-

type spectrometer further grinding has any advantage whatever.) Sufficient of the dry material is then placed on a watch glass and moistened with just enough 10 per cent Lucite lacquer to make the mixture of a creamy consistency. The lacquer is made by dissolving 10 g. of Lucite molding powder in 100 ml. ethyl acetate. It will be found convenient to have a small scoop of the correct size so as to measure quickly and reproducibly the desired amount of powder.

The sample with added Lucite lacquer is dried in an oven at about  $100^{\circ}$  and then crushed and lightly ground in a mortar. Next, the powder is placed in a rectangular mold together with a metal mount having a rectangular opening and subjected to a temperature that does not need to exceed  $160^{\circ}$  under a pressure that does not need to exceed  $200 \text{ kg/cm}^2$ . The pressure is applied after the sample has reached the desired temperature; and immediately thereafter the mold is cooled by a blast of air while the pressure is maintained.

For very soft materials, specimens of improved appearance will be obtained by using much less than the pressure mentioned above. Furthermore, with substances like the feldspars that have relatively low refractive indices, an irregular, mottled appearance may be produced unless the amount of Lucite is held to the bare minimum necessary for producing a coherent sample. The rectangular metal mounts made by the

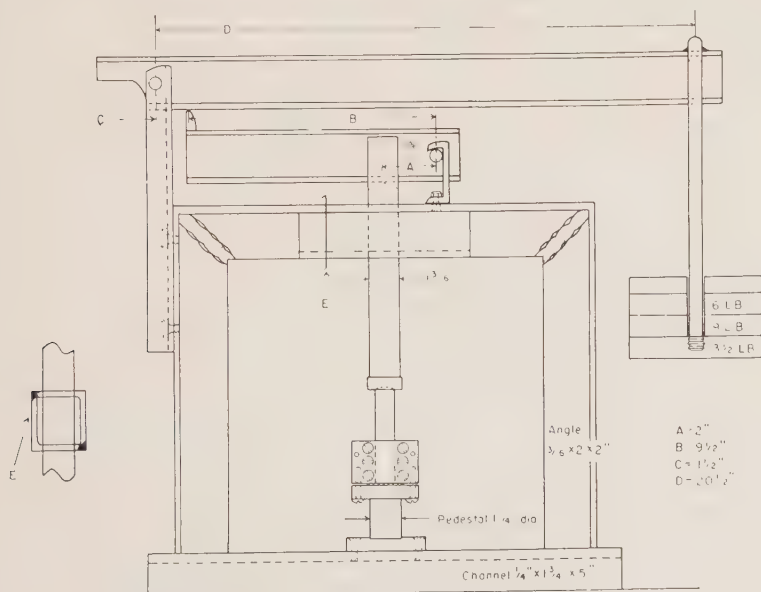


FIG. 1. Diagram showing in elevation the assembly of press and mold.

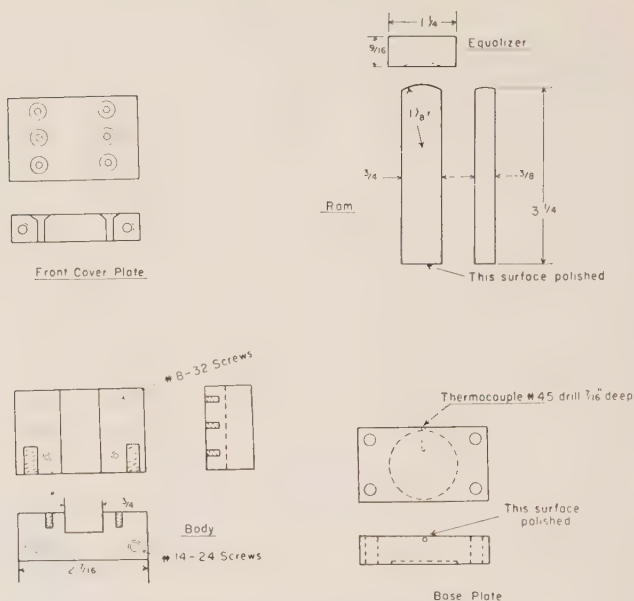


FIG. 2. Details of rectangular mold.

North American Philips Company are convenient. These holders are of aluminum alloy about 1.5 mm. thick having an opening of about 10 mm. by 20 mm.

With such holders and with the more common non-metallic substances about 0.2 g. will be sufficient to give a satisfactory specimen. If the supply of material is limited, smaller quantities will suffice; but an amount less than 70 mg. will yield only a thin and fragile layer in the usual mount. Specimens made according to the method described here need not contain more than 25 per cent by volume of the plastic.

Figures 1 and 2 respectively show the assembled press and the details of the rectangular mold. Our mold is made from scrap steel, the analysis of which we do not know, but Rockwell hardness tests indicate it to be probably a carbon tool steel with little or no temper. It has proved satisfactory with no tendency to gall or abrade. The body was made in two sections, dowelled and screwed together, and the ram fitted very closely so that little or no powder can slip through or into the moving surfaces. The lower end of the ram and top surfaces of the base plate are polished for smooth contact with faces of the sample. In order to assemble the mold, feed the ram through from the top until it projects about 1/16 inch beyond the bottom. Place the sample frame around the end of the



ram and clamp on the bottom plate. The ram may then be removed for charging the mold with sample material.

The electric heater is  $2\frac{3}{4}$  inches high and is otherwise shaped to the mold in a close but free fit, so that it will slide freely over the mold. The heater jacket or shell is made of 20 gauge sheet iron. The inner wall is wrapped with two layers of .025-inch asbestos paper (dampened), then wound with about 16 feet of No. 26 Nichrome wire, the resistance being approximately 42 ohms for convenient power input when used on a 110-volt circuit. After being wound, the wire is covered with two layers of asbestos and is then baked and assembled to the outer shell.

An air-blast cooler is convenient. This is made from brass tubing of the rectangular cross section,  $\frac{1}{4}$  inch  $\times$   $\frac{3}{8}$  inch, braced at the corners and measuring inside 2 inches  $\times$   $2\frac{5}{8}$  inches with a connection in the center for a rubber hose. A brass strip  $\frac{1}{8}$  inch  $\times$   $\frac{3}{4}$  inch running vertically and bolted to the base plate and top cross members provides the track for raising and lowering both heater and cooler, and a set screw mounted on a file handle is provided for each of these units as a means of locating the units where desired and holding them there.

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## SAPONITE NEAR MILFORD, UTAH

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### ABSTRACT

A small deposit of saponite has been found near Milford, Utah, which is apparently a result of hydrothermal replacement of a dolomitic limestone. Differential thermal analysis, optical data, chemical analysis, and x-ray analysis all confirm the identification of this mineral as saponite. Electron microscope pictures show this mineral to possess a sheet-like structure as expected.

### INTRODUCTION

A new deposit of saponite was found approximately seven miles southwest of Milford, Utah (see Fig. 1), in the Star Mining District. This district is in the most southern part of the area which was described by Butler (1) in 1913. The deposit was found in a small adit located near the Commonwealth mine (see Butler 1913). The area was actively mined for lead, silver, and copper in the 1870's and again around 1909. The present study began in the summer of 1951 for the purpose of positively identifying the mineral.

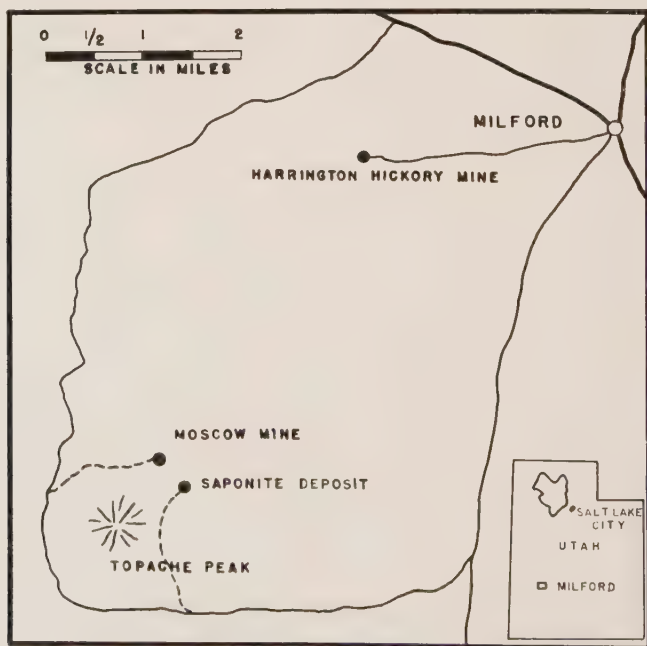


FIG. 1. Index map showing the location of the saponite deposit, Utah.

## GENERAL GEOLOGY

In the Star Mining District the rocks consist mainly of dolomitic limestones, quartzites, and an intrusive quartz monzonite. The beds strike generally north-south and dip approximately east at an angle of  $45^{\circ}$  to  $50^{\circ}$ . In general they are broken by a series of nearly parallel fissures which have an east-west strike with a very nearly vertical dip.

The saponite deposit is located in the Topache limestone thought by Butler to be of Mississippian age (1). It is found in a poorly defined vertical fissure of about 3 feet in width. In a hand specimen the clay is snow-white and porcelain-like in appearance, but is soft and has a greasy "feel." Associated with it is considerable siliceous material believed to be chalcedony.

## MINERALOGY OF THE CLAY

1. *Microscopic Properties*

The clay mineral was observed to be somewhat fibrous in its over-all appearance when viewed under low magnification, but under high magnification a more or less flaky appearance predominates. The indices of refraction of these flakes, using sodium light and immersion oils that were checked immediately with a refractometer, were found to be  $\alpha = 1.511$  and  $\gamma = 1.514$ . It should be pointed out that due to the small size of the flakes, these indices are at best only approximate.

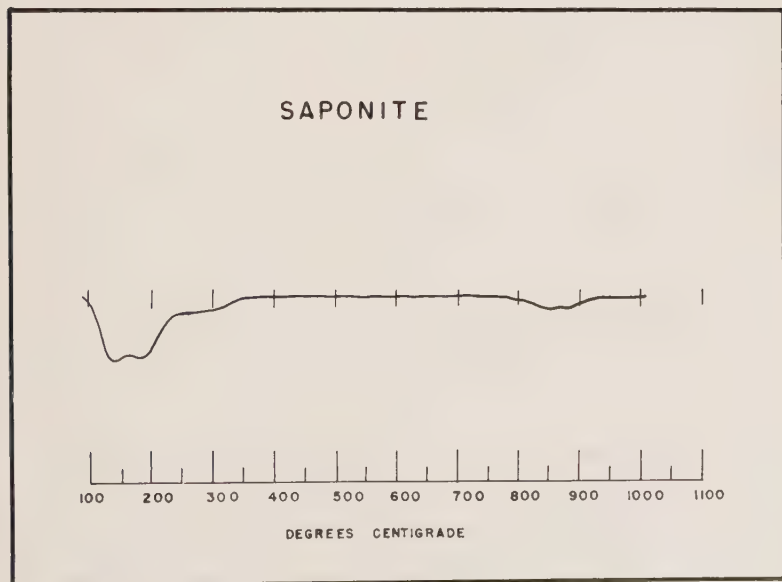


FIG. 2. Differential thermal analysis curve of saponite, Utah.

## 2. Differential Thermal Analysis

The apparatus used for this test consisted of a program controller constructed at the University of Utah to increase the temperature of the nichrome furnace at the rate of  $10^{\circ}$  C. per minute, and a six point, high speed, high sensitivity electronic recorder (Leeds and Northrup "Speedomax"), which recorded both furnace temperature and differential readings.

Figure 2 shows the curve obtained from differential thermal analysis of this mineral. The curves given by Kerr and Kulp (2) for saponite agree well with this curve except for the endothermic reaction at  $550^{\circ}$  C., which is absent in this case. The curve for saponite from Montreal, Quebec, as shown by Kerr and Kulp, also lacks this reaction.

## 3. Chemical Analysis

A spectrographic analysis run by Mr. Harold R. Bradford at the University of Utah gave very high percentages for magnesium and silicon, low percentages for aluminum, calcium and iron, and no lines for potassium, sodium, and a questionable line for lithium. A wet chemical analysis of hand picked material made by Mr. Walter Savournin at the University of Utah gave the following results:

TABLE 1. CHEMICAL ANALYSIS OF SAPONITE FROM MILFORD, UTAH

SiO <sub>2</sub>	50.01 Per cent
Al <sub>2</sub> O <sub>3</sub>	3.89
Fe <sub>2</sub> O <sub>3</sub>	0.21
CaO	1.31
MgO	25.61
TiO <sub>2</sub>	less than 0.04
H <sub>2</sub> O+	12.02
H <sub>2</sub> O—	7.28

Using these figures, the formula was calculated as  $17 \text{ MgO} \cdot \text{Al}_2\text{O}_3 \cdot 22\text{SiO}_2 \cdot 6\text{H}_2\text{O}(\text{CaO})$ . Calculations according to the method proposed by Ross and Hendricks (3) gave the structural formula as  $(\text{Al}_{0.04}\text{Fe}_{0.01}\text{Mg}_{2.85})(\text{Al}_{0.30}\text{Si}_{3.70})\text{O}_{10}(\text{OH})_2$ , which is in agreement with their formulas for saponite.

To form saponite's structure the alumina in the octahedral layer of the montmorillonite structure as given by Norton (4) would be almost completely replaced by magnesia. Therefore, the only difference in the structure for saponite would be a slightly thicker unit cell due to the substitution of the larger magnesium ions.



## 4. X-ray Analysis

An x-ray powder pattern was taken of the mineral after it had been exposed to room atmosphere for several days. The  $d$  values obtained are presented in Table 2. These values agree with the values given by MacEwan (5).

TABLE 2. X-RAY DATA OF SAPONITE, UTAH

Line Number	$d$ Values	Indices	$I/I_1$	Line Shape
1	16.6 13.0	001	1.0 VS	Vbr
2	4.94	001*	.2	Sh
3	4.51	110, 020‡	.8	Sh
4	3.70	?	.4	Br
5	3.21	002*	.1 V	
6	2.89	121*	.4	Br
7	2.58	130, 200‡	.5	Vbr <sup>1.</sup>
8	2.26	220, 040‡	.1 V	
9	2.06	003*	.1 V	
10	1.72	310, 150, ‡ 240	.3	Sh
11	1.52	330, 060‡	.9	Sh
12	1.31	260, 400‡	.5	Br
13	1.26	350, 170, ‡ 420	.1 V	
14	1.05	370, 280, ‡ 510	.1 V	
15	.99	190, 530, ‡ 460	.3	Br
16	.88	390, 600‡	.4	Sh

$d$  = Spacings in angstroms.

$I/I_1$  = Compared in tensities

V = Just visible

VS = Very strong

Vbr = Very broad

Br = Broad

Sh = Sharp

1. = A sharp inner edge

\* = Indices from Winkler x-ray Identifications and Structure of Clay Minerals, p. 126 (1951).

‡ = Indices from D. M. C. MacEwan X-ray Identification and Structure of Clay Minerals (1951), Table facing p. 124.

The mineral was also heated up to 1000° F. in intervals of 100 degrees with an x-ray photograph being taken after each interval. One photograph was taken of the mineral after it had been allowed to hydrate by placing it in a sealed jar containing water. It is interesting to note that the  $hk0$  spacings, in agreement with the findings of MacEwan (5), did not change with hydration or dehydration, but that the  $00l$  spacings

changed distinctly. This shift should be kept in mind when evaluating x-ray patterns of minerals of the montmorillonite type. The broadness of the 00 $l$  reflection can well be explained by the thickness of the particles. Line broadening is first noticeable when the mean thickness is about 1000 Å and is very strongly effective when the mean thickness reaches 300 Å.

### 5. *Electron Micrographs*

Samples were prepared for observation under the electron microscope by placing about 10 grams of mineral which had not been crushed in 100 cc. of distilled water. The container was thoroughly shaken once a day for a week. It was hoped by this method that the particles would separate from each other without disintegrating. Preparing the sample in this manner proved highly successful.

Figure 3 is an electron micrograph which shows a booklet of saponite in which part of the book has slid off the main particle. Several thin sheets are clearly visible. The flaw in the lower left hand corner of each sheet proves that they are all from the same booklet. It is possible to

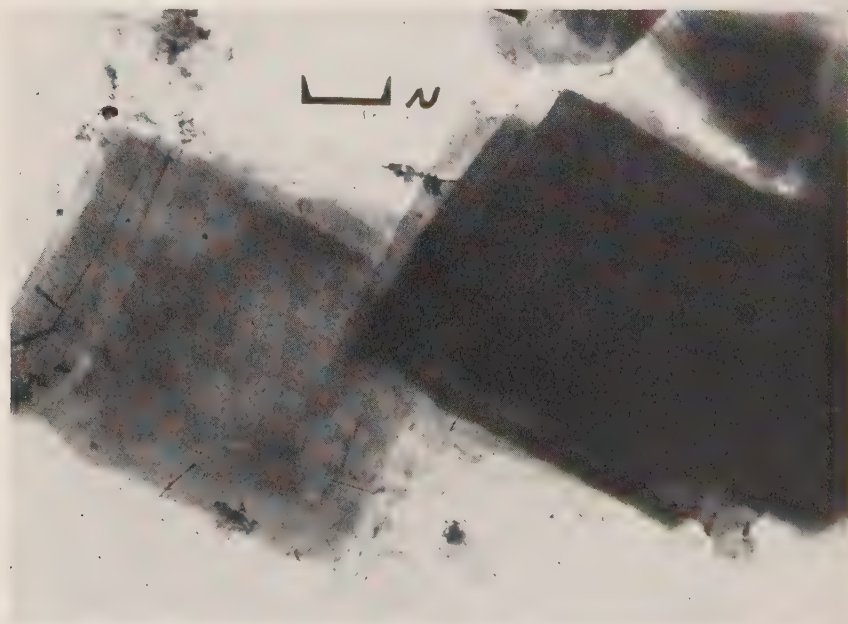


FIG. 3. Electron microscope photograph showing the book like structure of saponite. Notice the flaws in the sheets proving they are all from the same book. Notice the one or two very thin plates.

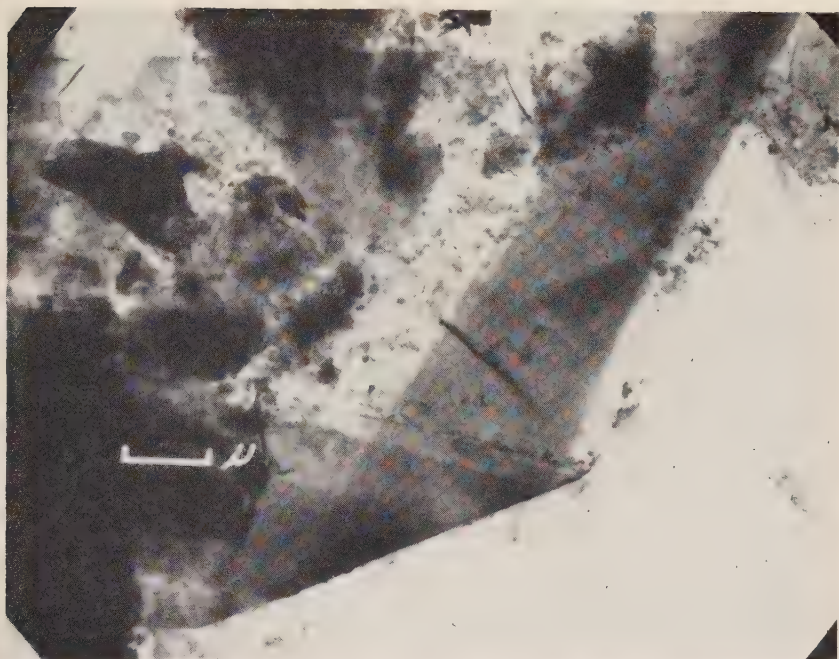


FIG. 4. Electron microscope picture showing a completely folded sheet. Notice that the individual sheets can be traced through the fold. Also note the one large thin sheet.

trace the outline of several of these thin sheets even though they overlie other sheets. Figure 4 shows a large multiple layer which has been folded. Tracing the individual sheets, it is possible to follow the edges through the fold. Also there is one large sheet which is approximately  $\frac{3}{4}$  as wide as the field, or about 12 microns in width. No tears or cracks are evident in the folded edge, which indicates that the sheet is pliable enough to withstand folding. The thinnest sheets have been torn in several places as evidenced by the small irregular particles in the field. The thinnest sheets seen in these figures are 50 Å and less in thickness,\* which is approaching the required thickness of a single unit cell.

The shapes of the particles probably have no significance in identifying this mineral in view of the fact that in all the photographs that have been observed no two particles possessed the same shape. Figure 5 shows

\* According to R. W. G. Wyckoff in his book *Electron Microscopy* (p. 52), the maximum thickness of objects that can be penetrated at an accelerating voltage of 50 KV is 2000 Å. Using this figure, the accelerating voltage used and the number of sheets observed on top of each other, the number 50 Å was calculated. At the present time experimental work is being done to determine the true thickness of these plates.



FIG. 5. Electron microscope photograph showing the sheets of saponite crinkled and bent. Most of the particles observed were of this type.

the most commonly observed particles. These particles are thin sheets which have been twisted and folded so that a wavy appearance resulted. Some of this effect no doubt is due to the incomplete dispersion of the particles. When a sample was prepared by the method previously described, except for the addition of three drops of sodium silicate, the sample expanded within a short time so that it was approximately 12 times its original volume. No individual particles were evident by visual inspection after this expansion and the mass became jelly-like in character. The expanded particles when viewed through the electron microscope were very thin and considerably smaller than the untreated particles. As is evident in Fig. 6 very few particles did not expand.

#### ORIGIN OF THE SAPONITE

The saponite occurs in dolomitic limestone as veins in a zone about three feet in width. In these veins are found nodules composed of a core of dolomitic limestone with a layer of pure white crystallized calcite surrounded by saponite (see Fig. 7). The dolomitic limestone cores are often partially replaced by veins and patches of chalcedony and saponite.



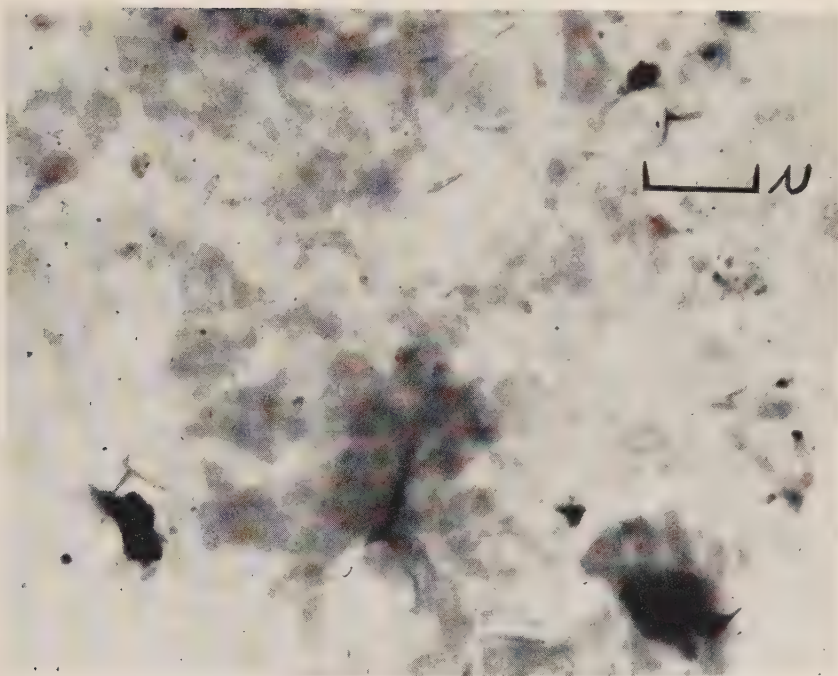


FIG. 6. Electron microscope photograph of the dispersed saponite. Notice that the particles have become very thin and small and are difficult to distinguish.

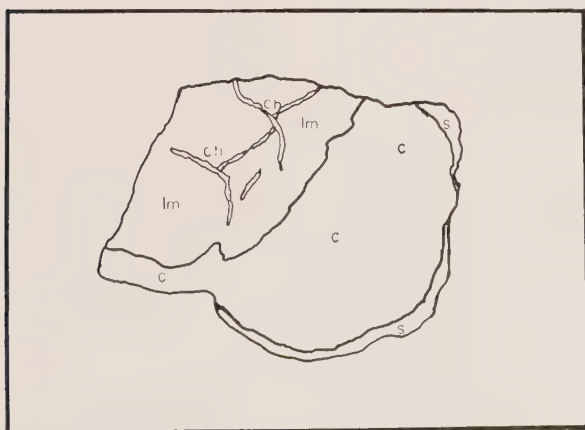


FIG. 7. Line drawing of a polished surface of a nodule showing the relation of limestone (*lm*), calcite (*C*), chalcedony (*Ch*), and saponite (*S*).



Microscopic veinlets of chalcedony, and rarely veinlets of saponite, are observed in the calcite. Occasionally small wheat-size grains of chalcedony are found within the saponite.

Silica-bearing hydrothermal solutions are believed to have formed this mineral by using the magnesium of the dolomitic limestone and depositing it as saponite, with the calcium of the limestone forming the calcite. The small amount of alumina could have been supplied by the clay impurities in the dolomitic limestone.

#### ACKNOWLEDGMENTS

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## CRYSTAL STRUCTURE OF MAGNESIUM-VERMICULITE

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### ABSTRACT

A single-crystal  $x$ -ray analysis of Mg-vermiculite has located the interlamellar water molecules and exchangeable cations in definite positions with respect to the adjacent silicate layer surfaces. The exchangeable cation sites lie in a plane midway between silicate layers. The water network consists of two sheets, each arranged in a distorted hexagonal pattern. The water molecule sites are determined by the surface configuration of the silicate layers, each water being linked by a hydrogen bond to a single oxygen in the silicate layer surface. Weak hydrogen-bonding operates within individual water sheets. The water sheets are held together by the exchangeable cations, around which the water molecules tend to form hydration shells. The importance of the hydration behaviour of the cations rather than direct electrostatic interaction between cations and silicate layer surfaces in locating the cations is emphasized.

A regular distortion of the surface oxygen network of the silicate layers from the ideal hexagonal form is revealed by the diffraction data.

Vermiculite must be regarded as a true clay mineral since it is formed in the clay fractions of certain soils (Walker, 1950*a*). The characteristic properties of this mineral, such as high cation exchange capacity (Walker, 1947, 1949; Barshad, 1948), the ability to form complexes with organic substances (Walker, 1950*b*; Barshad, 1952) and a variable interlamellar distance depending on the exchangeable cation present and the humidity of the sample (Barshad, 1949; Milne and Walker, 1950; Walker, 1951), bear a striking resemblance to those of montmorillonite. This similarity, moreover, extends to their structures which, in so far as they are known, consist of complex silicate layers interleaved with layers of water molecules carrying exchangeable cations.

The detailed structure analysis of montmorillonite has been prevented mainly by the small grain size of the particles, and the configuration of the interlamellar water molecules and exchangeable cations in this mineral is not well understood. Mg-vermiculite, on the other hand, is available in large flakes (derived by the hydrothermal alteration of biotite and phlogopite) which are suitable for investigation by single-crystal  $x$ -ray methods. Vermiculite therefore seems to offer a promising line of approach to the general problem of the configuration of water molecules and exchangeable cations in the vicinity of layer silicate surfaces, and the present investigation was undertaken primarily with this aim in view. From the analysis, certain new features of the internal structure of the silicate layers and of the relationships between adjacent

silicate layers have appeared which are not in accord with previous views.

Earlier workers on the crystal structure of vermiculite (Gruner, 1934, 1939; Hendricks and Jefferson, 1938*a*) were in general agreement regarding the structure of the silicate layers and their relative disposition when viewed along the *b* axis, but neither obtained experimental evidence of the structure of the interlamellar water layers. Hendricks and Jefferson (1938*b*) proposed a hypothetical structure for the interlamellar water which was based partly on Bernal and Fowler's (1933) concept of the tetrahedral charge distribution of a water molecule in liquid water. Neither Gruner nor Hendricks and Jefferson were aware, at the time of their work, of the high cation exchange capacity of vermiculite, and more complete information on the dehydration characteristics of the mineral has since become available.

#### EXPERIMENTAL

The vermiculite used came from Kenya and was obtained through the courtesy of Mr. G. E. Howling of the Imperial Institute, London. The sample has a cation exchange capacity of 130 milliequivalents per 100 grams, all the exchange positions being occupied by  $Mg^{2+}$  ions. The chemical analysis (Table 1) shows no alkalis, indicating that no interleaved layers of mica are present. Small sections, free from cracks, were cut from cleavage flakes and gently pressed flat to minimize any distortion arising from the cutting operation. The approximate dimensions of the crystals used in obtaining the diffraction data were  $0.5 \times 0.2 \times 0.2$  mm.

Rotation films indicated that, for the three  $9.18 \text{ \AA}$  axes disposed at

TABLE 1. CHEMICAL ANALYSIS AND FORMULA OF KENYA VERMICULITE  
(air dry)

	Wt. %
$SiO_2$	34.04
$Al_2O_3$	15.37
$Fe_2O_3$	8.01
$MgO$	22.58
$CaO$	0.00
$Na_2O$	0.00
$K_2O$	0.00
$H_2O$	19.93
	99.93
$Mg^{2+}_{0.32}$	
↑	
$(Mg_{2.26}Fe^{+3}_{0.48}Al_{0.16})(Al_{1.28}Si_{2.72})O_{10}(OH)_2 \cdot 4.32 H_2O$	
Or	
$[C_3D_4O_{10}(OH)_2]^{-0.64} [4.32H_2O \cdot 0.32 Mg^{2+}]^{+0.64}$	

120° with respect to each other in the cleavage plane, the sharp  $3n$ -layer line spectra coincide in position and intensity, but that marked differences occur in the intensity distribution of the  $(3n \pm 1)$ -layer spectra. Oscillation films disclosed only one axis with equivalence of both sharp and diffuse  $+n$ - and  $-n$ -layer spectra. The crystal system is therefore monoclinic. Moving films of the zero, first, second and third layers about the  $b$ -axis were recorded on an equi-inclination Weissenberg goniometer (Mathieson, 1951). The cell constants obtained are in good agreement with those of Gruner (1934), but differ from those of Hendricks and Jefferson (1938a) in the selection of the  $\beta$ -angle.

	Gruner	Hendricks and Jefferson	Mathieson and Walker
$a$	5.3	5.33	5.33 Å
$b$	9.2	9.18	9.18 Å
$c$	28.57–28.77	28.85	28.90 Å
$\beta$	97°09'	93°15'	97°
Space Group	$Cc$ or $C2/c$	$Cn$ or $C2/n^*$	$Cc$

\* Reported as  $Cc$  or  $C2/c$ .

Comparison of the  $h0l$  and  $h3l$  moving-film photographs shows that, for  $\beta = 97^\circ$ , the unit cell corresponding to the sharp  $k = 3n$  spectra is centered on the  $C$  face and contains one silicate layer ( $c = 14.45$  Å). Inclusion of the diffuse  $k \pm 3n$  spectra requires a doubling of the  $c$  axis

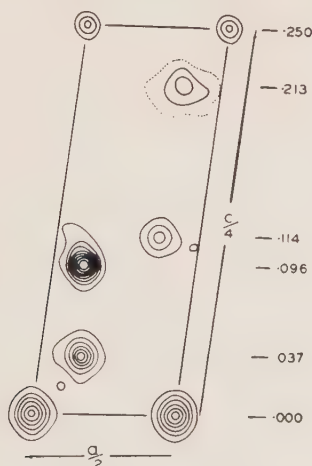


FIG. 1. (a) Electron-density distribution on (010) for Mg-vermiculite. Contours for octahedral (C) atoms at intervals of  $4e \cdot \text{\AA}^{-2}$ , for tetrahedral (D) atoms and oxygens  $2e \cdot \text{\AA}^{-2}$ , for  $\text{H}_2\text{O}$  and exchangeable  $\text{Mg}^{2+}$  at  $1e \cdot \text{\AA}^{-2}$ . The dotted line represents the  $1e \cdot \text{\AA}^{-2}$  level. Distances parallel to the  $z$ -axis are indicated at the right-hand side of the diagram.

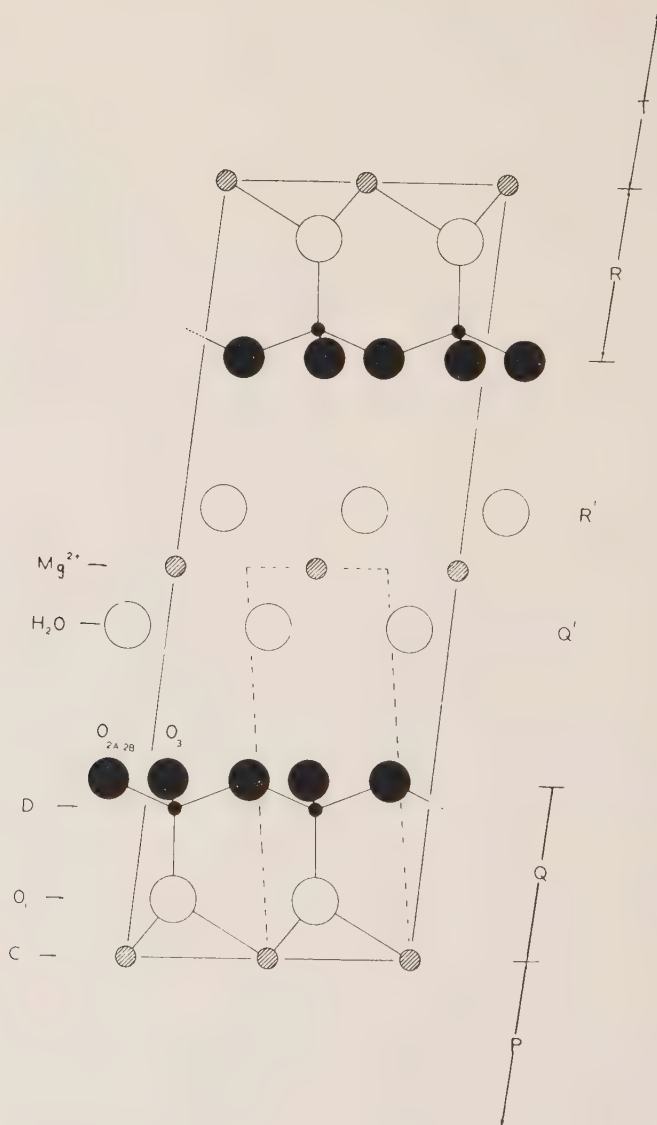


FIG. 1. (b) The corresponding crystal structure of Mg-vermiculite projected on (010).  $P$ ,  $Q$ ,  $R$  and  $T$  refer to silicate half-layers;  $Q'$  and  $R'$ , sheets of water molecules;  $C$ , (Mg, Fe, Al) octahedrally-coordinated atoms;  $D$ , (Al, Si) tetrahedrally-coordinated atoms;  $O_1$ , oxygens and hydroxyls at  $z=0.037$ ;  $O_{2A}$ ,  $O_{2B}$ ,  $O_3$ , oxygens at  $z=0.114$ ;  $H_2O$ , interlamellar water molecules;  $Mg^{2+}$ , interlamellar cations. An area,  $(a/2) \times (c/4)$ , of the unit cell selected by Hendricks and Jefferson (1938a) is indicated by the broken lines.



(i.e.,  $c = 28.90 \text{ \AA}$ ). The general reflexions,  $hkl$ , occur only for  $h+k=2n$ . The space group for the structure, based on this value of  $\beta$ , is  $Cc$  or  $C2/c$ . If the value  $\beta = 93.5^\circ$  is used, the  $k=3n$  spectra are accounted for by a cell centered on all faces and containing two silicate layers. The correct space group for this unit cell is either  $Cn$  or  $C2/n$ , not  $Cc$  or  $C2/c$  as reported by Hendricks and Jefferson (1938a). The relation of their unit cell to the one selected in the present study is shown in Fig. 1b.

Intensity data for the  $00l$ ,  $h0l$  and  $h3l$  reflexions were recorded on the Weissenberg goniometer using filtered Mo radiation. For each set of reflexions, two packs, each of four films interleaved with tin-foil to increase the inter-film ratio, were exposed for 100 and 5 hours respectively. Intensity estimations were made by comparison with a set of standards obtained by timed exposures of a selected individual reflexion oscillated over a small angular range. For the diffuse reflexions, a crystal was chosen which gave some indication of diffraction maxima rather than the very diffuse streaks produced by most specimens. Filtered Cu radiation was utilised to provide adequate resolution of the peaks. The distribution of the  $02l$  reflexions was measured by means of a Leeds-Northrop recording microphotometer and corrected for film background (Fig. 2).

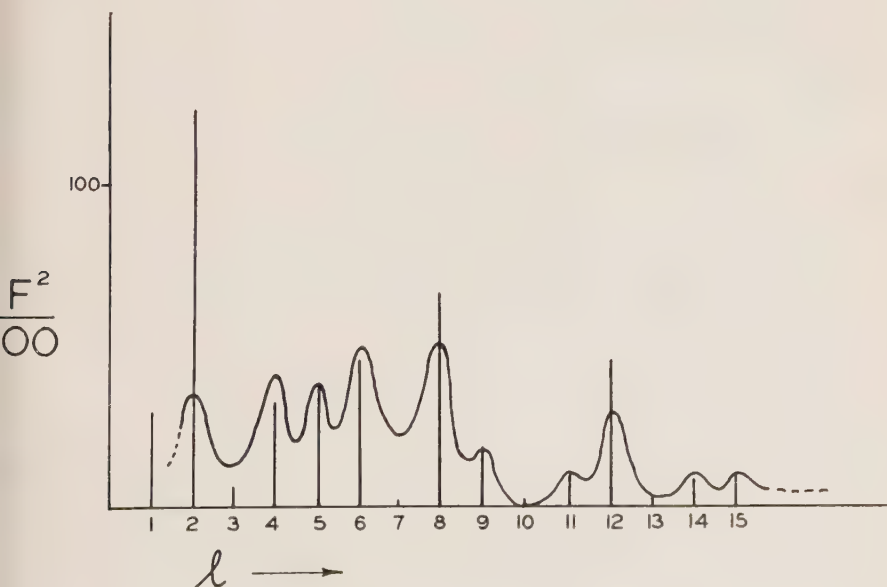


FIG. 2. Photometer curve of the  $02l$  diffuse reflexions, corrected for film background. The calculated values for the mean positional arrangement of structures  $q$  and  $r$  are superimposed for comparison.

The  $F^2$  values of the  $00l$ ,  $h0l$  and  $h3l$  reflexions were obtained after correction for polarization and Lorentz factors by graphical means (Kaan and Cole, 1949). The structure amplitudes were later placed on an approximately absolute scale by correlation with the calculated values. For the calculation of structure amplitudes, the scattering factors for  $Mg^{2+}$  and  $Si^{4+}$  (Internationale Tabellen, 1935) were loaded by the respective isomorphous replacements indicated by the formula based on a chemical analysis of the sample (Table 1). The temperature factor,  $B = 1.2 \times 10^{-16}$  cm.<sup>2</sup>, was determined by plotting  $\log \Sigma |F_c| / \Sigma |F_o|$  against  $\sin^2 \theta$  in six equal ranges of  $\sin^2 \theta$  from 0.0 to 0.6 (Wilson, 1942). Fourier summations were carried out on three-figure Beevers-Lipson strips, the  $a$  and  $c$  axes being subdivided into 30 and 120 parts respectively. The electron-density map was drawn from contour values derived from interpolated sections.

Because of the frequent association of chlorite and vermiculite as mixed-layer structures, it was necessary to be certain that the specimen under investigation contained no interleaved chloritic material, although previous examination of the total sample had indicated its purity. The crystal from which the  $h0l$  and  $h3l$  data had been obtained was therefore converted to Li-vermiculite, and the  $00l$  reflexions rephotographed. A prolonged exposure showed the characteristic 12.2 Å basal reflexion of Li-vermiculite with a regular series of higher orders (Milne and Walker, *loc. cit.*). No sign of a 14 Å reflexion, such as would be produced by the presence of chlorite, could be found. On this basis, it is estimated that the specimen contained less than 0.5 per cent of chlorite layers.

### STRUCTURE ANALYSIS

Initial attempts at a unidimensional synthesis of this vermiculite were reported by Milne and Walker (1951), and an interim statement of results in the present investigation was made in 1952 (Mathieson and Walker).

#### 1. *Electron-density distribution normal to (001)*

For the preliminary synthesis, the signs of the  $00l$  structure amplitudes of Mg-vermiculite recorded up to the 62nd order were computed from the contributions of the atoms of the silicate layer alone. Omission of the water molecules and exchangeable cations from the calculations ensured that no assumptions would be involved in their location. Apart from the distribution corresponding to the silicate layer, the synthesis disclosed two additional peaks, one at  $z = 0.214$  and a smaller one at  $z = 0.250$ .

A unidimensional synthesis of the same crystal after conversion to

Sr-vermiculite reproduced the electron-density curve of the Mg-vermiculite except for the peak at  $z=0.25$ , which was substantially increased in size. The exchangeable cations are therefore located midway between the silicate layers. The water molecules are arranged on either side of the central cations in sheets which lie at a distance of 2.83 Å from the surface oxygens of the adjacent silicate layers.

Recalculation of the structure amplitudes, taking into account the contributions of the water molecules and exchangeable cations, gave reliability factors,

$$R = \frac{\sum |F_o - F_c|}{\sum |F_o|},$$

of 0.14 and 0.22 for Mg- and Sr-vermiculite respectively (Table 2).

## 2. Projection along the *b*-axis

Since the approximate configuration of the silicate layer is known, this portion of the structure can be disposed in the unit cell by consideration of the structure amplitudes for the reflexions 20,12 to 204 (see Table 2). With the approximate  $x$  parameters derived by this means and taking the  $z$  parameters from the line synthesis, a preliminary set of structure amplitudes for the  $h0l$  reflexions was calculated. Again the contributions of the water molecules and exchangeable cations were omitted initially so that no assumptions would be involved in their location. The resultant electron-density contour map provided more accurate parameters for the atoms of the silicate layer, and located the water molecules at  $x=0.142$ ,  $z=0.213$  and the exchangeable cations at  $x=0.0$ ,  $z=0.250$ . Recalculation of the structure amplitudes for all constituents of the unit cell necessitated changes in sign for a few minor terms only, and a subsequent synthesis produced the electron-density distribution shown in Fig. 1*a*. The corresponding diagram of the crystal structure is given in Fig. 1*b*. The final reliability factor for  $h0l$  reflexions was 0.15.

## 3. The Crystal Structure in Three Dimensions

Initially, deductions arising from the sharp  $k=3n$  spectra will be considered and the results referred to a sub-cell containing a single silicate layer ( $c=14.45$  Å).

From the equivalence of the  $[010]$ ,  $[310]$  and  $[3\bar{1}0]$  axes, it is evident that the projections of the crystal structure along  $[310]$  and  $[3\bar{1}0]$  will be identical with that along  $[010]$  (Fig. 1*b*). The three projections can therefore be combined to give the complete structure if the coordination values of the atoms are also taken into account. The crystal structure of half a silicate layer (e.g., Q, Fig. 1*b*) viewed normal to  $(001)$  is shown

TABLE 2. COMPARISON OF OBSERVED AND CALCULATED STRUCTURE AMPLITUDES FOR (a) Sr-VERMICULITE AND (b) Mg-VERMICULITE. NOTE THAT  $F(20l) = F(13, l+2)$ : SLIGHT DIFFERENCES IN THE CALCULATED VALUES FOR THESE SETS OF REFLEXIONS ARE DUE TO APPROXIMATIONS IN THE ATOMIC PARAMETERS AND SCATTERING FACTORS

(a)			(b)								
00 <i>l</i>	<i>F</i> <sub>obs.</sub>	<i>F</i> <sub>calc.</sub>	00 <i>l</i>	<i>F</i> <sub>obs.</sub>	<i>F</i> <sub>calc.</sub>	<i>h</i> 0 <i>l</i>	<i>F</i> <sub>obs.</sub>	<i>F</i> <sub>calc.</sub>	<i>h</i> 0 <i>l</i>	<i>F</i> <sub>obs.</sub>	<i>F</i> <sub>calc.</sub>
002	—	298	002	282	305	20, 58	<25	6	20, $\overline{56}$	<23	—11
004	—	78	004	62	39	20, 56	<24	21	20, $\overline{58}$	<24	—11
006	—	-93	006	110	-72	20, 54	<23	20	20, $\overline{60}$	<25	—11
008	219	168	008	211	213	20, 52	<22	6	20, $\overline{62}$	<27	—11
00, 10	344	336	00, 10	342	313	20, 50	<22	-1			
00, 12	55	21	00, 12	60	-67	20, 48	<21	-12	40, 52	<26	-11
00, 14	185	-114	00, 14	105	-79	20, 46	<20	14	40, 50	<26	-11
00, 16	63	-76	00, 16	74	-95	20, 44	37	28	40, 48	29	-11
00, 18	<20	4	00, 18	86	124	20, 42	32	39	40, 46	46	-11
00, 20	209	224	00, 20	167	161	20, 40	<19	17	40, 44	23	-11
00, 22	66	69	00, 22	84	91	20, 38	<18	-9	40, 42	<24	-11
00, 24	150	150	00, 24	121	121	20, 36	68	70	40, 40	<23	-11
00, 26	52	41	00, 26	42	34	20, 34	100	92	40, 38	46	-11
00, 28	54	88	00, 28	83	110	20, 32	105	122	40, 36	63	-11
00, 30	65	85	00, 30	68	69	20, 30	16	6	40, 34	35	-11
00, 32	76	87	00, 32	107	108	20, 28	16	-20	40, 32	<20	-2
00, 34	94	95	00, 34	61	61	20, 26	16	31	40, 30	<20	-11
00, 36	64	44	00, 36	<20	2	20, 24	105	87	40, 28	<19	-11
00, 38	39	-30	00, 38	38	-19	20, 22	79	109	40, 26	52	53
00, 40	<26	-9	00, 40	<22	-11	20, 20	37	-57	40, 24	52	47
00, 42	<27	-13	00, 42	41	59	20, 18	63	-48	40, 22	29	37
00, 44	61	76	00, 44	39	33	20, 16	37	23	40, 20	29	42
			00, 46	<24	-7	20, 14	173	173	40, 18	46	20
			00, 48	<24	-11	20, 12	263	257	40, 16	150	142
			00, 50	<25	20	20, 10	105	87	40, 14	162	143
			00, 52	54	76	208	178	189	40, 12	144	152
			00, 54	39	33	206	116	81	40, 10	<16	-8
			00, 56	<26	9	204	304	300	408	16	-33
			00, 58	<26	-7	202	205	212	406	63	81
			00, 60	<27	9	200	136	127	404	167	134
			00, 62	27	23	202	16	20	402	58	94
						204	147	173	400	110	-126
						206	32	51	402	87	-69
						208	100	72	404	127	122
						20, 10	147	175	406	225	223
						20, 12	58	41	408	167	178
						20, 14	47	-40	40, 10	<16	-30
						20, 16	152	163	40, 12	58	73
						20, 18	236	203	40, 14	127	132
						20, 20	294	281	40, 16	196	206
						20, 22	63	37	40, 18	75	77

TABLE 2—(continued)

(b)											
00l	$F_{obs.}$	$F_{calc.}$	00l	$F_{obs.}$	$F_{calc.}$	h0l	$F_{obs.}$	$F_{calc.}$	h0l	$F_{obs.}$	$F_{calc.}$
						20, $\overline{24}$	<16	1	40, $\overline{20}$	<18	-23
						20, $\overline{26}$	16	30	40, $\overline{22}$	29	-7
						20, $\overline{28}$	105	100	40, $\overline{24}$	<18	9
						20, $\overline{30}$	89	98	40, $\overline{26}$	52	61
						20, $\overline{32}$	<17	-24	40, $\overline{28}$	58	31
						20, $\overline{34}$	52	-30	40, $\overline{30}$	17	26
						20, $\overline{36}$	<18	-11	40, $\overline{32}$	<20	18
						20, $\overline{38}$	37	57	40, $\overline{34}$	<20	13
						20, $\overline{40}$	73	70	40, $\overline{36}$	63	68
						20, $\overline{42}$	37	25	40, $\overline{38}$	81	66
						20, $\overline{44}$	26	31	40, $\overline{40}$	81	76
						20, $\overline{46}$	<20	11	40, $\overline{42}$	<22	8
						20, $\overline{48}$	47	54	40, $\overline{44}$	<23	-5
						20, $\overline{50}$	47	36	40, $\overline{46}$	<23	17
						20, $\overline{52}$	37	28	40, $\overline{48}$	40	32
						20, $\overline{54}$	<22	7	40, $\overline{50}$	<24	27

h0l	$F_{obs.}$	$F_{calc.}$	h0l	$F_{obs.}$	$F_{calc.}$	h0l	$F_{obs.}$	$F_{calc.}$	h3l	$F_{obs.}$	$F_{calc.}$
0, $\overline{52}$	<25	-24	80, 20	<23	5	12, $\overline{08}$	47	24	13, 46	47	53
0, $\overline{54}$	<25	-18	80, 18	<23	19	12, 0, $\overline{10}$	<30	14	13, 44	<27	11
0, $\overline{56}$	<26	8	80, 16	<22	4	12, 0, $\overline{12}$	<30	1	13, 42	21	30
0, $\overline{58}$	23	29	80, 14	<22	-9	12, 0, $\overline{14}$	<30	2	13, 40	28	23
0, $\overline{60}$	<27	22	80, 12	<22	-2	12, 0, $\overline{16}$	<30	11	13, 38	68	67
			80, 10	42	30	12, 0, 18	<30	10	13, 36	46	55
0, 42	<23	-7	808	79	70				13, 34	26	-12
0, 40	<23	-7	806	37	42				13, 32	17	-28
0, 38	<22	30	804	<21	20				13, 30	48	-24
0, 36	<22	12	802	<21	4				13, 28	88	97
0, 34	<21	-10	800	63	54				13, 26	98	96
0, 32	<20	-26	802	74	74				13, 24	15	27
0, 30	20	19	804	32	31				13, 22	<23	2
0, 28	76	72	806	26	-24				13, 20	62	38
0, 26	50	51	808	16	-32				13, 18	209	271
0, 24	<19	27	80, $\overline{10}$	<20	27				13, 16	204	196
0, 22	<19	-5	80, $\overline{12}$	48	51				13, 14	139	157
0, 20	50	57	80, $\overline{14}$	<20	19				13, 12	45	-40
0, 18	81	77	80, $\overline{16}$	<20	-6				13, 10	49	40
0, 16	66	55	80, $\overline{18}$	<21	2				138	171	180
0, 14	<18	-2	80, $\overline{20}$	37	56				136	120	74
0, 12	35	-34	80, $\overline{22}$	63	60				134	32	49
0, 10	<17	14	80, $\overline{24}$	58	47				132	206	169



TABLE 2—(continued)

$h0l$	$F_{obs.}$	$F_{calc.}$	$h0l$	$F_{obs.}$	$F_{calc.}$	$h0l$	$F_{obs.}$	$F_{calc.}$	$h3l$	$F_{obs.}$	$F_{calc.}$
608	45	35	80,26	<22	17				130	<20	
606	60	56	80,28	<22	3				132	141	1
604	35	36	80,30	<22	18				134	234	2
602	<17	10	80,32	<22	13				136	313	2
600	35	43	80,34	<23	17				138	141	
602	71	62	80,36	<23	0				13,10	174	1
604	141	142	80,38	<23	-7				13,12	113	
606	96	91	80,40	<24	-7				13,14	227	22
608	35	43	80,42	<24	7				13,16	179	11
60,10	20	-18	80,44	<25	32				13,18	32	
60,12	<18	21	80,46	<25	19				13,20	49	-
60,14	76	90	80,48	<26	13				13,22	45	-
60,16	66	54							13,24	76	14
60,18	<19	-11	10,0,14	<26	21				13,26	95	
60,20	50	-57	10,0,12	47	37				13,28	21	
60,22	<19	12	10,0,10	26	24				13,30	<23	-
60,24	86	94	10,08	<25	13				13,32	<24	
60,26	81	75	10,06	<24	14				13,34	101	1
60,28	30	34	10,04	<24	9				13,36	112	
60,30	<20	-9	10,02	<23	16				13,38	63	
60,32	30	52	10,00	<23	6				13,40	<25	
60,34	60	67	10,02	<23	6				13,42	<26	
60,36	55	53	10,04	<22	0				13,44	29	
60,38	<21	7	10,06	<22	-8				13,46	30	
60,40	<21	-15	10,08	<22	10				13,48	<28	
60,42	<22	7	10,0,10	36	28						
60,44	<22	9	10,0,12	42	43				33,40	<28	
60,46	<23	14	10,0,14	<23	15				33,38	<27	
60,48	<24	3	10,0,16	<23	5				33,36	<27	-
60,50	<24	0	10,0,18	<24	18				33,34	<26	-
60,52	<25	10	10,0,20	36	41				33,32	23	2
60,54	<26	11	10,0,22	26	32				33,30	78	9
			10,0,24	<25	-7				33,28	68	7
80,32	<27	34	10,0,26	<25	-21				33,26	51	9
80,30	<26	30	10,0,28	<26	0				33,24	<24	-1
80,28	<25	27	10,0,30	<26	19				33,22	73	7
80,26	<25	11	10,0,32	<27	15				33,20	132	11
80,24	<24	-1							33,18	136	11
80,22	<24	12	12,06	<30	19				33,16	<23	1

TABLE 2—(continued)

<i>hkl</i>	<i>F</i> <sub>obs.</sub>	<i>F</i> <sub>calc.</sub>	<i>hkl</i>	<i>F</i> <sub>obs.</sub>	<i>F</i> <sub>calc.</sub>	<i>02l</i>	<i>F</i> <sub>calc.</sub>	$\frac{F^2 \text{ calc.}}{100}$
33,14	70	82	53,30	44	41	021	54	29
33,12	<22	7	53,32	46	70	022	111	123
33,10	90	60	53,34	<28	—9	023	—25	6
338	139	134	53,36	<29	—25	024	56	31
336	65	50	53,38	<29	—19	025	—61	37
334	<22	—8	53,40	<30	45	026	—67	45
332	107	104	53,42	<30	58	027	—11	1
330	207	140				028	—81	66
332	315	304	73,20	<28	13	029	43	18
334	175	140	73,18	<27	1	02,10	10	1
336	82	102	73,16	<26	26	02,11	30	9
338	<22	—5	73,14	76	79	02,12	67	45
33,10	58	53	73,12	76	73	02,13	—15	2
33,12	116	143	73,10	34	37	02,14	29	8
33,14	84	59	738	<25	—1	02,15	—31	10
33,16	—23	4	736	<25	9			
33,18	62	—88	734	45	50			
33,20	<23	24	732	56	38			
33,22	130	135	730	<25	20			
33,24	117	122	732	<25	—35			
33,26	62	79	734	<25	—4			
33,28	<24	—15	736	50	41			
33,30	36	71	738	78	72			
33,32	68	89	73,10	62	68			
33,34	78	99	73,12	<25	18			
33,36	23	24	73,14	<25	27			
33,38	<25	—30	73,16	<26	45			
33,40	<26	0	73,18	64	72			
33,42	<26	16	73,20	56	55			
33,44	<27	33	73,22	<26	—2			
33,46	<27	4	73,24	<27	—13			
			73,26	<28	—12			
53,32	55	58	73,28	34	36			
53,30	54	71	73,30	34	29			
53,28	<26	23	73,32	<30	11			
53,26	<25	—15	73,34	<32	—6			
53,24	<25	13						
53,22	43	39	936	<28	22			
53,20	67	72	934	<28	30			
53,18	<24	—21	932	<27	8			
53,16	40	—35	930	<27	—5			
53,14	<24	—4	932	<26	21			
53,12	109	102	934	53	47			
53,10	135	133	936	53	56			

TABLE 2—(continued)

$h3l$	$F_{obs.}$	$F_{calc.}$	$h3l$	$F_{obs.}$	$F_{calc.}$	$02l$	$F_{calc.}$	$\frac{F^2_{calc.}}{100}$
538	52	51	938	<25	31			
536	<24	27	93,10	<25	-1			
534	43	40	93,12	<25	-12			
532	143	134	93,14	<25	-3			
530	142	130	93,16	<25	7			
532	49	34	93,18	39	25			
534	<24	-4	93,20	<26	31			
536	43	-47	93,22	<27	12			
538	43	58	93,24	<28	10			
53,10	68	53						
53,12	58	70						
53,14	<24	16						
53,16	<25	4						
53,18	47	64						
53,20	96	102						
53,22	111	127						
53,24	50	35						
53,26	<26	-2						
53,28	<27	13						

in Fig. 3a. On the basis of the diffraction data considered so far, the arrangement of the water molecule sites and the exchangeable cation sites ( $m_1$ ,  $m_2$ ,  $m_3$ ) can be represented diagrammatically by the view (normal to (001) and projected on the base of the cell) of the layer  $Q'R'$  (Fig. 3b). The chemical data indicate that the water molecule and exchangeable cation sites are approximately two-thirds and one-ninth filled respectively. Calculation of the  $h3l$  structure amplitudes for this structure gives satisfactory agreement with the observed values (Table 2) establishing its essential correctness ( $R=0.17$ ). Table 3 lists the atomic coordinates; the alternative sets of values ( $a$ ) and ( $b$ ), given for the  $x$  and  $y$  coordinates of the water molecule sites, will be discussed at a later stage.

The structure of the silicate layer which emerges is very similar to that put forward by previous workers, although greater detail is revealed than has hitherto been obtained for layer silicate structures, largely because of increased precision of intensity estimation combined with refinement by successive Fourier syntheses. The bond lengths are as

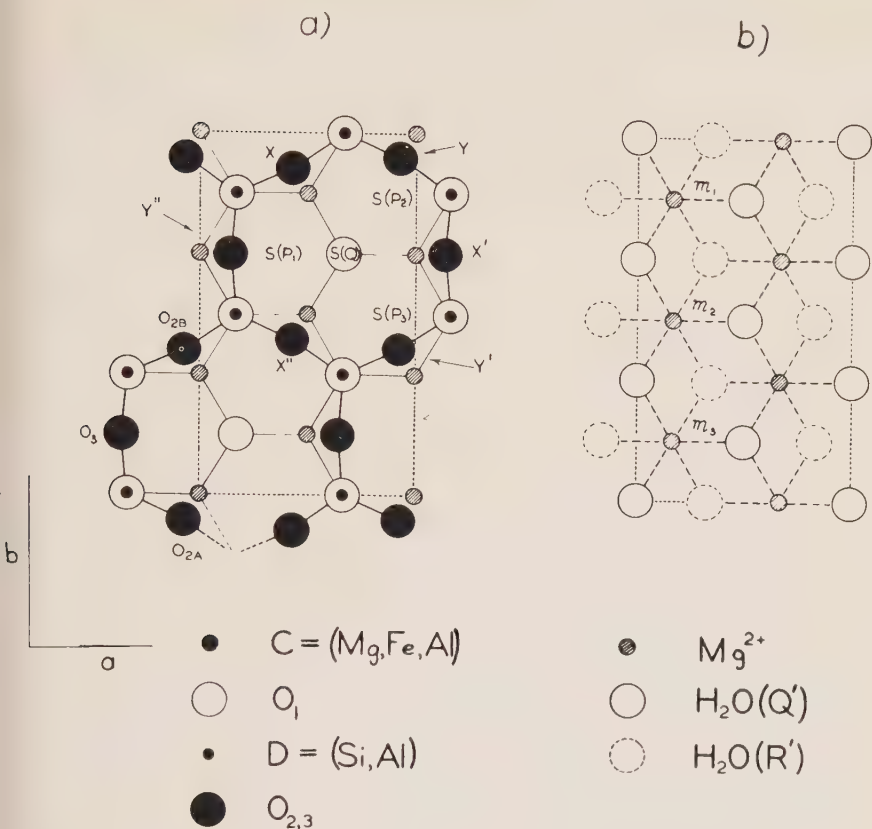


FIG. 3. (a) Section of the crystal structure of Mg-vermiculite from  $z=0.0$  to  $0.114$  (i.e. half-layer  $Q$ ) projected on the base of the cell normal to  $(001)$ . (b) Section of the crystal structure from  $z=0.213$  to  $0.287$ , i.e., sites of the water molecules in sheets  $Q'$  and  $R'$  and sites of exchangeable  $\text{Mg}^{2+}$  cations.

follows:  $C^*-O_1$ ,  $2.06 \text{ \AA}$ ;  $D^*-O_1$ ,  $1.69 \text{ \AA}$ ; and  $D-O_{2A}$ ,  $D-O_{2B}$ ,  $D-O_3$ ,  $1.62 \text{ \AA}$  each (probable error,  $\pm 0.02 \text{ \AA}$ ). The oxygen atom  $O_1$  is linked tetrahedrally with one  $D$  (or hydrogen) and three  $C$  atoms, the angle  $COD$  being  $109^\circ$ , whereas  $O_{2A}$ ,  $O_{2B}$  and  $O_3$  are each linked to two  $D$  atoms with a  $DOD$  angle of  $142^\circ$ . If  $r(\text{Si}^{4+})$  is taken as  $0.41 \text{ \AA}$ , then  $r(O_1)$  is  $1.28 \text{ \AA}$  and  $r(O_2, O_3)$ ,  $1.21 \text{ \AA}$ . The relationship between ionic radius and coordination number (Wells, 1950, p. 71) gives the ratio of two- to

\* For convenience,  $C$  will be used to represent the  $(\text{Mg}, \text{Fe}, \text{Al})$  atoms in octahedral positions at the centre of the silicate layer; and  $D$ , the  $(\text{Si}, \text{Al})$  tetrahedrally-coordinated atoms near the surface. The key to the oxygen atoms is given in Figs. 1b and 3a.

four-coordinated radii as  $(2/4)^{\frac{1}{2}} = 0.92$  and the observed ratio, 0.95, is therefore of the correct order.

The contour map clearly indicates a distortion in the surface oxygen sheet of the silicate layer. The hexagon of oxygen atoms  $XYX'Y'X''Y''$  (Fig. 3a) is not regular but consists of two interpenetrating isosceles triangles of oxygens  $XX'X''$  and  $YY'Y''$  with sides 4.35 and 4.84 Å respectively. The corresponding distance in a regular hexagon would be 4.59 Å. The distortion is accounted for by a rotation through  $5\frac{1}{2}^\circ$  of the triad of oxygens above each *D* atom. Since the change from the ideal hexagonal to the distorted configuration would involve differences in *D*-O bond lengths of less than 0.01 Å, it is improbable that the distortion arises from inability to accommodate a regular hexagonal oxygen network in the cross-sectional area of the unit cell (i.e.,  $9.18 \times 5.33$  Å). The distortion is more readily explained on the basis of electrostatic forces within the silicate layer, if it is assumed that a residual negative charge on each surface oxygen interacts with a residual positive charge on the octahedral *C* atom lying below and to one side (viewed normal to (001)). The electrostatic forces thus invoked are correctly disposed to produce a torque in the required direction (Fig. 3a), the angular movement being limited by the over-riding factor of the *D*-O distance.

At this juncture, it is appropriate to consider the approximate spatial relationships of the interlamellar water and exchangeable cation sites to an adjacent silicate layer. It will be assumed for the moment that the water molecules all lie at the centre of the peak of the electron-density distribution (Fig. 1a), although the possibility that this is merely a mean position will be suggested later. The sites for the water molecules lie directly over *C* atoms and those for the exchangeable cations over *D* atoms (or hydroxyls) of the silicate layer. This latter observation indicates an important point with regard to the relationship of adjacent silicate layers, namely the collinearity of (i) atoms *O*<sub>1</sub> and *D* (or *H*) of the half-layer *Q*, (ii) the exchangeable cation site, and (iii) atoms *D* (or *H*) and *O*<sub>1</sub> of the half-layer *R* (Fig. 1b).

The structure of complete silicate layers and the method of stacking of the layers can be deduced from a consideration of the  $k \pm 3n$  spectra. The water molecules and exchangeable cations were omitted from the calculations since they can only have a minor influence on the structure amplitudes. The 02*l* reflexions were employed in testing the various possible structures.

In discussing the silicate layers, it is convenient to focus attention on the site at the centre of the distorted hexagon of surface oxygen atoms. Let this site be *S*(*Z*) where *Z* refers to the half-layer *P*, *Q*, *R* or *T*. Then the half-layers *P* and *Q* combine to form a single layer and *S*(*Q*) can be associated with *S*(*P*<sub>1</sub>), *S*(*P*<sub>2</sub>) or *S*(*P*<sub>3</sub>) (Fig. 3a). These three possible



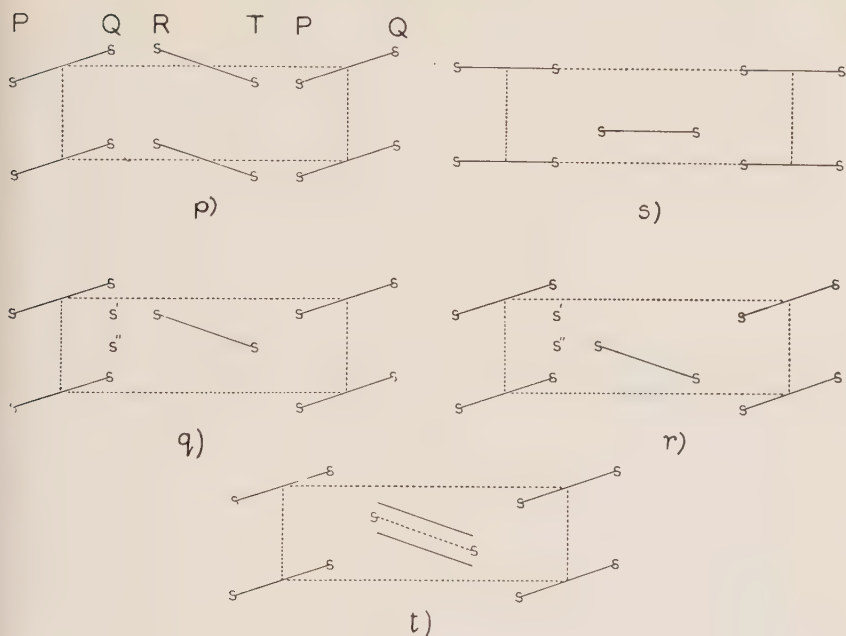


FIG. 4. Schematic presentation of the possible stacking sequences in Mg-vermiculite. (*p*) (*q*) and (*r*) *N*-type silicate layer displaced with respect to an *M*-type layer by 0,  $b/3$  and  $2b/3$  respectively. (*s*) *L*-type layers displaced by  $b/3$  (or  $2b/3$ ). (*t*) Positional mean of structures *q* and *r*.

layer structures correspond to the *L*, *M* and *N* types of layers referred to by Brindley, Oughton and Robinson (1950) in their study of the chlorites. The three types of silicate layer have individual symmetry  $C2/m$  and are indistinguishable until the *b* axis of the crystal is defined by the mutual relations of the stacked layers. When a second complete silicate layer *RT* is added, there are twelve possible ways of grouping *P*, *Q*, *R* and *T*. The existence of a glide operation *c* reduces the possibilities to four, illustrated diagrammatically in Fig. 4 *p*, *q*, *r* and *s*. As Hendricks and Jefferson (1938*a*) have shown, none of these arrangements yields structure amplitudes in agreement with the observed intensity distribution. However, structures *q* and *r* give some measure of agreement for reflections  $02l$  where  $l=2n$ , and a structure with the unit *RT* located at the centre of the projection area (Fig. 4 *t*) gives a very close agreement with the observed distribution of intensities for the  $02l$  spectra. Table 2 lists the amplitudes calculated for this arrangement, and Fig. 2 shows the  $F^2$  values superimposed on the photometer curve.

The solution obtained in this way is completely contrary to all the evidence of the sub-cell analysis, according to which the site *S*(*R*) should

lie over the site  $S(Q)$  or displaced therefrom by  $\pm b/6$  in this projection (i.e.,  $S'$  or  $S''$ , Fig. 4). Furthermore, a unique structure for vermiculite would be implied by structure  $l$  and therefore no explanation for the occurrence of the diffuse spectra would be possible. The deductions from both the sharp and diffuse spectra can be reconciled only if it is assumed that structures  $q$  and  $r$  are equally probable and that structure  $p$  does not occur. In other words, the stacking of the silicate layers in vermiculite corresponds to  $M$ -type layers alternating with  $V$ -type layers. The layers, however, can only be arranged so that site  $S(R)$  is not in line with site  $S(Q)$  nor  $S(P)$  with  $S(T)$ . The resultant crystal structure can be represented for the purposes of calculation by the average structure  $l$  (Fig. 4). The mutual relationships of the surface oxygens of the silicate half-layers  $Q$  and  $R$  are illustrated in Fig. 5.

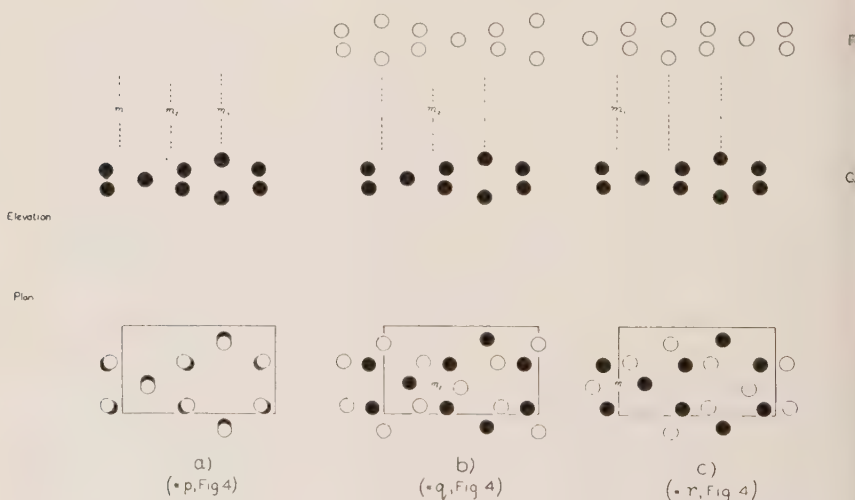


FIG. 5. Plan and elevation of the relationship between the surface oxygen network of half-layers  $Q$  and  $R$ .  $m_1$ ,  $m_2$ ,  $m_3$  are sites for exchangeable cations as deduced from the subcell analysis, and are defined by their relationship to the half-layer  $Q$ . Projections are normal to (001).

#### 4. Location of the Sites of the Water Molecules

In the  $b$ -axis projection (Fig. 1a), the contours of the water molecules tend to be drawn out parallel to the  $a$  axis and there is evidence of a tail in the negative  $x$ -direction. As a result, the peak height is lower than would be expected for 2.16 water molecules (Table 1).

At first sight, the shape of the water peak might seem to be due to high thermal energy in the water molecules, producing movement mainly parallel to (001) because of restrictions imposed by the environment.

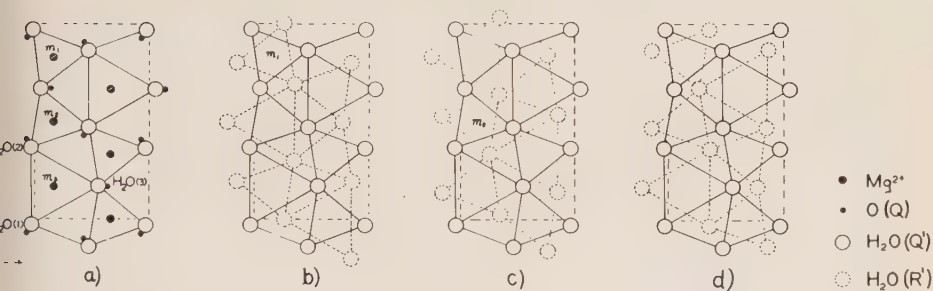


FIG. 6. (a) Diagram of the relation of the distorted water network  $Q'$  to the surface oxygen network of the half-layer  $Q$ . Cation sites  $m_1, m_2, m_3$  are also shown. This diagram illustrates the tendency of each water molecule to approach more closely to its associated surface oxygen. (b), (c), (d) The three ways of combining two sheets of water sites, the relative shifts being  $b/3, 2b/3$  and  $0$  respectively. (d) is excluded by the data. The sites  $m_1$  and  $m_2$  with suitable undistorted octahedral environment for the  $Mg^{2+}$  cations are shown.

This explanation, however, does not account for the "tail" on the water molecule peak and, in order to do so, it must be assumed that the asymmetric electron-density distribution corresponds to regular displacements of the water molecules from the mean position (Table 3, column (a)). In this event, one would expect such displacements to be related to the adjacent oxygen network and, in a triad of water molecules ( $H_2O(1), H_2O(2), H_2O(3)$ , Fig. 6a) over a surface oxygen hexagon, each member of the triad would be displaced equally towards or away from its related oxygen, i.e., the triad would expand or contract. If it expands, there must be concomitant rotations of unexpanded triads in opposite directions around  $m_1$  and  $m_2$  (Fig. 6a), and the corresponding electron-density distribution viewed along the  $b$ -axis would develop a tail in the negative  $x$ -direction such as is indicated in the contour map (Fig. 1a)\*. The distortion of the water sheet produced in this way results in a closer approach of each water molecule to its related oxygen atom. The relevant atomic parameters are listed in column (b), Table 3.

Since each water sheet is located by its relation to the adjacent silicate layer surface, a complete water layer (consisting of two sheets of water molecules) can only be formed by superimposing the water sheets in the two ways permitted by the stacking of the silicate layers. These two arrangements correspond to relative shifts of  $b/3$  and  $2b/3$ , and are illustrated in Fig. 6 b and c respectively. Inspection shows that the two arrangements are equivalent and, in both, the relative displacements of the water molecules maintain normal  $H_2O-H_2O$  approach distances be-

\* The displacement of the water molecules from their ideal hexagonal positions cannot be determined accurately from the shape of the electron-density distribution but is probably of the order of  $0.2 \text{ \AA}$ .

TABLE 3. ATOMIC PARAMETERS FOR MG-VERMICULITE

Atom		<i>x</i>		<i>y</i>		<i>z</i>
<i>C</i>		0		0		0
<i>C</i>		0		0.333		0
<i>C</i>		0		0.667		0
<i>O</i> <sub>1</sub>		0.358		0		0.037
<i>O</i> <sub>1</sub>		0.358		0.333		0.037
<i>O</i> <sub>1</sub>		0.358		0.667		0.037
<i>D</i>		0.397		0		0.096
<i>D</i>		0.397		0.333		0.096
<i>O</i> <sub>2A</sub>		0.147		0.404		0.114
<i>O</i> <sub>2B</sub>		0.147		0.929		0.114
<i>O</i> <sub>3</sub>		0.434		0.167		0.114
		( <i>a</i> )	( <i>b</i> )	( <i>a</i> )	( <i>b</i> )	
H <sub>2</sub> O	site (1)	0.142	0.160	0	-0.019	0.213
	site (2)	0.142	0.160	0.333	0.352	0.213
	site (3)	0.142	0.105	0.667	0.667	0.213
Mg <sup>2+</sup>	site <i>m</i> <sub>1</sub>	0.500		0		0.250
	site <i>m</i> <sub>2</sub>	0.500		0.333		0.250
	site <i>m</i> <sub>3</sub>	0.500		0.667		0.250

tween the sheets. The method of stacking the water sheets illustrated in Fig. 6 *d* is excluded, since it could only exist if structure *p* (Fig. 4) were permissible. The nonoccurrence of structure *p* is perhaps connected with the fact that such a water-layer arrangement would involve very close approach of the water molecules between the sheets.

### 5. The Structure of the Interlamellar Region

From the previous sections, it is evident that the arrangement of the *sites* of the interlamellar cations and water molecules closely resembles that of the octahedral *C* atoms and *O*<sub>1</sub> atoms, respectively, of the silicate layers although, in the former, the water sites are displaced somewhat from the regular octahedral arrangement. According to the chemical data, however, only a proportion of the interlamellar water and cation sites are occupied at one time.

Since each water site is equivalent with respect to the associated oxygen of the silicate layer surface, any grouping of the water molecules will be due to the influence of the cations. The importance of the cations in this respect is suggested by the variation of the interlamellar spacing produced when other cations are substituted for the Mg (Milne and Walker, 1950). A certain amount of evidence has been adduced by others to show that Mg ions tend to attract octahedral hydration shells in

water solution (e.g. Bernal and Fowler, 1933), and such octahedral grouping is known to occur in crystal hydrates such as magnesium chlorate hexahydrate (West, 1935). It is reasonable to suppose, therefore, that there will be a tendency for the exchangeable Mg ions in vermiculite to form hydration shells.

Examination of the two equivalent water-layer structures in Fig. 6 *b* and *c* reveals that in each case only one of the three *m* cation sites provides *undistorted* octahedral coordination for a cation (*m*<sub>1</sub> in Fig. 6 *b* or *m*<sub>2</sub> in Fig. 6 *c*). The Mg<sup>2+</sup>—H<sub>2</sub>O distance of 2.06 Å involved, moreover, is in agreement with that observed in magnesium benzene sulphonate hexahydrate (Broomhead and Nicol, 1948) which is the most accurate value available. All other sites have an asymmetric environment of water molecules, consisting of an unexpanded triad in one sheet and an expanded triad in the other. Equal Mg<sup>2+</sup>—H<sub>2</sub>O distances at these sites could only be maintained by displacing the cations 0.06 Å from the central plane, which is beyond the limit of error of measurement from the contour map (Fig. 1*a*). The resultant equilibrium Mg<sup>2+</sup>—H<sub>2</sub>O distance, moreover, would be 2.20 Å. It is almost certainly true, therefore, that the cations will occupy *m*<sub>1</sub> or *m*<sub>2</sub> sites depending on the stacking of the adjacent silicate layers.

The degree of ordering of the water molecules and cations within the available sites can now be considered further. The possibility of order *within an individual water-cation layer* is not excluded by the diffraction data since mutual ordering of these layers would be necessary for their structure to be revealed by the data. A case for the existence of long-range order of this nature, not directly deducible from the diffraction evidence, has been convincingly put forward in recent studies of hollandite (Byström, 1951) and psilomelane (Wadsley, 1952). So far, we have shown that the exchangeable cations in vermiculite lie in a plane midway between adjacent silicate layers, and that their lateral disposition is related to the adjacent water sheets. There are, however, only enough cations to fill one in three of the available *m*<sub>1</sub> (or *m*<sub>2</sub>) sites.

If it is assumed that the cations, due to mutual repulsion effects, tend to distribute themselves uniformly throughout the available sites, a hexagonal distribution of the type shown in Fig. 7 is obtained. Six water molecules can be arranged around and in contact with a Mg ion at a *u* site, a triad in the upper and a triad in the lower sheet, forming the single hydration shell already postulated. In the lower sheet, additional waters can be added in groups of three around unoccupied sites *v* or *w*. The water molecule sites around *w* in this sheet will be closer to the cations than those around *v*, so that the former sites will be preferred if it is assumed that the tendency of the cations to form single hydration



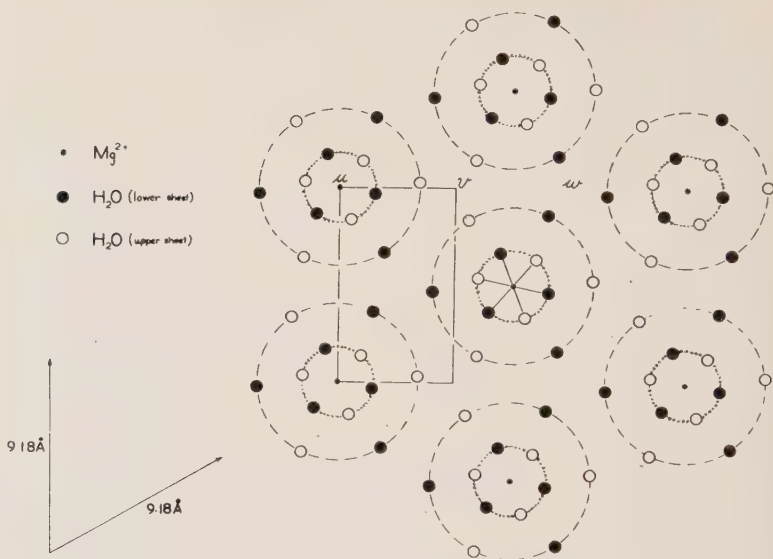


FIG. 7. Diagram of the proposed structure of the interlamellar region. Sites  $u$ ,  $v$ ,  $w$ , are all of type  $m_1$  (or all of type  $m_2$ ).

shells indicates a similar although less-marked tendency to form second shells.\* In the upper sheet, sites around  $v$  will be preferred to those around  $w$  since the former are in this case closer to the cations. Such an arrangement allows each Mg ion to have around it an almost complete double shell of water molecules, and the linking-up of these double shells produces layers in which two out of every three water sites are occupied. In this arrangement of the water layers, there are holes corresponding to triads of vacant water sites, each triad being equidistant from three cations with their associated double hydration shells. These sites may tend to be vacant (at normal temperature and pressure) because a water molecule in such a site would be in the third hydration shells of two cations and hence less stable than those associated with a single cation.

The types of bonding operating within the interlamellar region can now be considered with the aid of measurements taken from the contour map (Fig. 1a). If each water molecule (e.g.  $H_2O(A)$ , Fig. 8) is attached to its related oxygen by means of a hydrogen bond ( $H_2O-O$ , 2.84 Å), the second hydrogen can be directed towards another water molecule ( $H_2O(B)$ ) in the same sheet, so that the  $H-O-H$  angle is  $104\frac{1}{2}^\circ$  in agreement with the spectroscopically measured value (Darling and

\* Support for the assumption is provided by evidence of the occurrence of  $Mg^{2+} \cdot 12H_2O$  groups in salt solutions (Sidgwick, 1950, p. 237), and by the high energy of hydration of Mg cations.

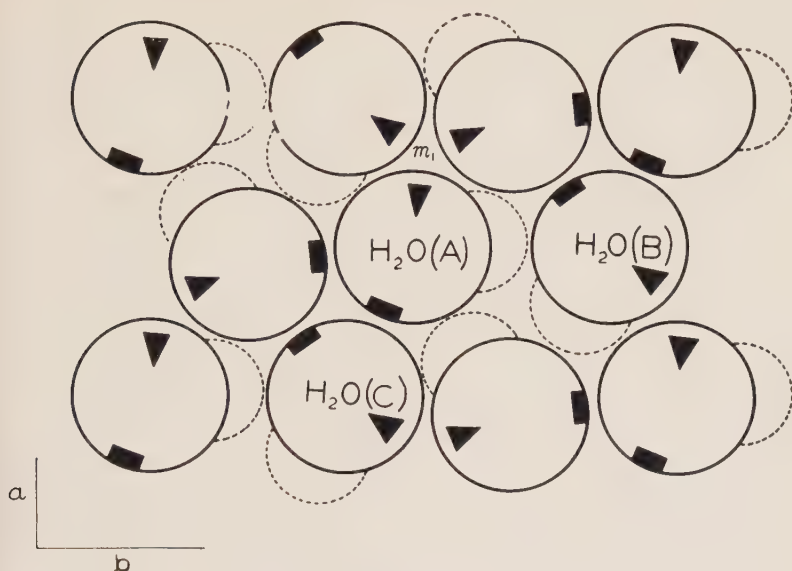


FIG. 8. Suggested system of weak hydrogen bonding illustrated for a single water sheet assuming all water molecule sites filled. Oxygens of water molecules represented by full circles, packing radius (Rees, 1949) to scale; hydrogens linked to silicate surface oxygens not shown; hydrogens in the plane of the water sheet represented by broken semi-circles; negative poles represented by black triangles and rectangles, triangles indicate greater displacement of charge from the plane of the sheet than rectangles.

Dennison, 1940). The second hydrogen lies in the plane of the water sheet but the two negative poles of the water molecule are slightly displaced therefrom, one directed towards the Mg ion site and the other pointing in the general direction of a third water molecule site ( $\text{H}_2\text{O}(C)$ ).

Since the intermolecular distances within a water sheet are either 3.06 Å or 3.40 Å, it is clear that normal hydrogen bonding does not operate at these points, and that the interactions are of a less specific nature. They may be regarded as *weak* hydrogen bonds and, as a consequence, a rigid application of tetrahedral linkages need not apply. In the vicinity of a cation site, the observed intermolecular distance within a water sheet of 3.06 Å is greater than the value calculated for a perfectly regular octahedron of waters around a Mg ion (viz. 2.92 Å). The triads of water molecules associated with a cation are expanded slightly in the plane of the sheets so as to allow each water to approach its associated oxygen. As a consequence, the cations sink slightly into the water triads in both sheets and the intersheet distance of the water molecules is reduced to 2.88 Å so as to preserve the Mg— $\text{H}_2\text{O}$  distance of 2.06 Å. The intersheet distance of 2.88 Å is uniform throughout the water

layers, which provides a possible further reason for the occurrence of triads of vacant water sites associated with the unoccupied *v* and *w* cation sites where the binding effect of the cations is absent. The observed intermolecular distance in an expanded triad of waters, viz. 3.40 Å, is consistent with the model if packing radii are assumed (Rees, 1949).

The idealised arrangement proposed above (Fig. 7) gives two out of three water sites occupied, which is approximately the required ratio, and the ratio of exchangeable cations to waters in the network is one in twelve which is close to the value indicated by the chemical formula (1:13.5). This model may therefore be closely approximated in the Mg-vermiculite under examination. A highly-ordered arrangement of the interlamellar region is not inconsistent with the observed ease of cation replacement (Milne and Walker, 1950), since normal diffusion processes can be expected to operate through vacant *u* sites and "interstitial" positions *v* and *w*, with concomitant adjustments in the water network. It is evident that the interlamellar region must be regarded as a dynamic system in which constant rearrangement and adjustment is taking place.

#### DISCUSSION

Detailed analysis has revealed a distortion in the surface oxygen network of the silicate layer in vermiculite. The explanation advanced is based on residual electrostatic forces acting between the central octahedrally-coordinated atoms and the surface oxygens of the silicate layers. Evidence for the existence of a distortion of the silicate layer in muscovite has been noted previously by Hendricks and Jefferson (1939), although in this case the data did not allow the nature of the distortion to be defined. Slight departures from regularity are therefore possible in silicate layers of this type.

The solution advanced for the stacking of the silicate layers, if valid, would impose certain limitations on similar minerals, and may therefore be applicable to the stacking sequences in talc and pyrophyllite. No satisfactory explanation for the  $0kl$ ,  $k \pm 3n$  spectra has so far been deduced for these minerals. The observed structure amplitudes of the  $02l$  reflexions in vermiculite have been shown to correspond closely to the values calculated not for *q* or *r* (Fig. 4) separately, but for the positional mean of these two possible arrangements. It is necessary to point out, however, that the results of the interpretation here placed on the diffuse reflexions cannot be reconciled with the theoretical treatment of disorder for cobalt in the region  $\alpha \sim \frac{1}{2}$  (Wilson, 1949, p. 70), which is analogous to the present case and predicts the almost complete disappearance of the diffuse regions. The discrepancy may arise from the failure of the theoretical

treatment to take into account the apparent increase in symmetry of the structure in the region  $\alpha \sim \frac{1}{2}$ .

The main interest attached to the structure analysis lies in the light which it throws on the organization of the interlamellar region. The water-cation network consists of two sheets of distorted hexagonally-linked water molecule sites with the cations occupying definite positions in the midway plane. Although the chemical data require that only one-third of cation sites ( $m_1$  or  $m_2$ ), and two-thirds of water sites should be occupied, it has been shown that a random filling of the available sites is not a necessary consequence of the data and, in the vermiculite under examination, a highly-ordered arrangement of the water molecules and cations is probable. In vermiculites which carry a higher population of exchangeable cations (Barshad, 1950, 1952), the idealised structure would be realised to a lesser extent, since  $u$ ,  $v$  and  $w$  sites would be expected to be partly occupied by cations and the distribution of the water molecules in the available sites would be altered accordingly. Nevertheless, grouping of the water molecules around the cations and a tendency towards a uniform distribution of the cations are to be expected in all similar minerals if the assumptions made above are valid.

It has usually been assumed, in considering the interaction of exchangeable cations with the planar surfaces of layer silicates, that the former are located by reactive spots of negative charge on the silicate layer surfaces arising from individual isomorphous substitutions. The present structure analysis has located the interlamellar  $Mg^{2+}$  ions in a plane midway between silicate layers. In this plane, the potential energy due to electrostatic forces can be shown by calculation to be effectively uniform, if the high frequency of isomorphous substitution is taken into account.\* The distribution of the cations in the midway plane is therefore not related to any *directive* effect of the electrostatic forces. The importance of the hydration behaviour of the cations in this respect is attested by the relationship between the cations and the water molecules as revealed by the diffraction data, and also by the observed spontaneous rehydration of partially-dehydrated Mg-vermiculite (Walker, 1949). In the interlamellar region, the water molecules within a sheet are linked by weak hydrogen bonds and the individual water sheets are held together by the cations. The hydrogen bonds binding the water molecules to the oxygen atoms of the silicate surfaces are probably stabilised to some extent by the electrostatic forces acting through the water molecules. We may conclude that, in Mg-vermiculite, the location of the

\* In vermiculite, which is a trioctahedral mineral, the substitution of about one in three tetrahedral and about one in five octahedral atoms gives rise to negative and positive charges respectively. The result is a net negative charge acting at the silicate layer surfaces.



water molecule sites is determined by the surface configuration of the silicate layers, and that the mutual relationship of the water sheets in a single water-cation layer is determined by the requirement for octahedral coordination of the water molecules around the cations. The unimportance of direct electrostatic interaction between the cations and surface oxygens is consistent with the conclusion of Guggenheim and McGlashan (1951) that the influence of next-nearest neighbours is entirely negligible. Their result, however, does not invalidate the proposed tendency to the formation of second hydration shells around the interlamellar cations in Mg-vermiculite, since each water molecule is equivalent in position with respect to the adjacent silicate layer surface and dipole interaction between water molecules can therefore exert a significant influence.

The structure derived for the interlamellar region in Mg-vermiculite, even in its less specific form, excludes the hypothetical water-layer structure proposed by Hendricks and Jefferson (1938*b*) for vermiculite, and later modifications made to take account of the exchangeable cations (e.g. Hendricks, Nelson and Alexander, 1940). Furthermore, the structure observed does not appear to have been considered in any of the models proposed for the water-cation network of layer silicate minerals.

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# RELATIVE STABILITIES OF SOME SIMPLE SILICATES AS RELATED TO THE POLARIZATION OF THE OXYGEN IONS

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## ABSTRACT

The stabilities of simple silicates relative to the component oxides are discussed. Some general relationships between stability of these silicates and size and charge of the cations are pointed out. The larger the size of the noble gas type cation and/or the lower its charge, the more stable the silicate relative to the free oxides. This is explained in principle by a simple crystal model in which the oxygen ion becomes unsymmetrically polarized in the resultant field created by  $\text{Si}^{4+}$  and the metal ions. The model is put in a mathematical form, and it can be shown by the equation how the  $\text{Si}^{4+}-\text{O}^{2-}$  bonds become strengthened and the cation- $\text{O}^{2-}$  bonds weakened when the oxides react to form a silicate. The energy release combined with the strengthening of the  $\text{Si}^{4+}-\text{O}^{2-}$  links is usually larger than the energy absorption connected with the weakening of the cation- $\text{O}^{2-}$  links thus resulting in a net energy release when stable silicates form from the free oxides. This exothermic energy is related to the degree of polarization of the oxygen ion; the more polarized the oxygen the greater the energy release and the more stable the silicate relative to the free oxides.

The empirical relationship between degree of polymerization in silicates and the field strength of the cation is discussed.

Although many of the stability relationships can be accounted for in principle by employing the concept of polarization and contrapolarization of the oxygen ions, it is realized that the concept of resonance between ionic and covalent bindings also can explain the observations. The reason that the idea of polarization is made use of in this paper is that it can be put more readily in a mathematical form.

## INTRODUCTION

It is a known crystal chemical fact that the stability of silicates and other oxysalts is related to the polarizing power of the cations or their tendencies to form covalent bonds with oxygen (Goldschmidt, (1927), Cartledge (1951), Ramberg (1952).

One may say that a cation with strong polarizing power or pronounced tendency to make covalent bonds loosens the bonds within the oxyanions, like  $\text{SiO}_4^{4-}$ ,  $\text{PO}_4^{3-}$ ,  $\text{CO}_3^{2-}$ , etc., thus causing a splitting up of the oxysalts into the free oxides. Cations with small polarizing power allow strong bonds to form within the oxyanion and do therefore make stable silicates or other oxysalts. This phenomenon is very significant for the understanding of formation of silicates and deserves more attention from mineralogists and geochemists than it has hitherto attracted. Not only is the question whether an element can form silicates or not controlled by

TABLE 1

Li $\text{Li}_2\text{Si}_2\text{O}_5$	Be $\text{Be}_2\text{SiO}_4$	B none	C none
Na $\text{Na}_2\text{Si}_2\text{O}_5$	Mg $\text{MgSiO}_3$	Al $\text{Al}_2\text{OSiO}_4$	Si none (quartz)
K $\text{K}_2\text{Si}_4\text{O}_9$	Ca $\text{CaSiO}_3$	Sc $\text{Sc}_2\text{Si}_2\text{O}_7$	Ti none
Rb $\text{Rb}_2\text{Si}_4\text{O}_9$	Sr $\text{SrSiO}_3$	Y $\text{Y}_2\text{Si}_2\text{O}_7$	Zr $\text{ZrSiO}_4$
Cs $\text{Cs}_2\text{Si}_4\text{O}_9$	Ba $\text{BaSi}_2\text{O}_5$	lanthanides ?	Hf $\text{HfSiO}_4$
X	Ra ?	Ac ?	Th $\text{ThSiO}_4$

TABLE 2

Ion	Ionic potential	Type of silicates				
		ortho	pyro	meta	phyllo	$\text{Si}_4\text{O}_9^{2-}$ -type
$\text{C}^{4+}$	26	none	none	none	none	none
$\text{B}^{3+}$	15	none	none	none	none	none
$\text{Ti}^{4+}$	6.25	none	none	none	none	none
$\text{Be}^{2+}$	5.88	$\text{Be}_2\text{SiO}_4$	none	none	none	none
$\text{Al}^{3+}$	5.26	$\text{Al}_2\text{OSiO}_4$	none	none	none	none
$\text{Zr}^{4+}$	4.60	$\text{ZrSiO}_4$	none	none	none	none
$\text{Th}^{4+}$	3.64	$\text{ThSiO}_4$	none	none	none	none
$\text{Sc}^{3+}$	3.61	—	$\text{Sc}_2\text{Si}_2\text{O}_7$	none	none	none
$\text{Y}^{3+}$	2.83	—	$\text{Y}_2\text{Si}_2\text{O}_7$	none	none	none
$\text{Mg}^{2+}$	2.56	$\text{Mg}_2\text{SiO}_4$	—	$\text{MgSiO}_3$	none	none
$\text{Ca}^{2+}$	1.89	$\text{Ca}_2\text{SiO}_4$	—	$\text{CaSiO}_3$	none	none
$\text{Sr}^{2+}$	1.57	$\text{Sr}_2\text{SiO}_4$	—	$\text{SrSiO}_3$	none	none
$\text{Ba}^{2+}$	1.40	$\text{Ba}_2\text{SiO}_4$	—	$\text{BaSiO}_3$	$\text{BaSi}_2\text{O}_5$	none
$\text{Li}^+$	1.28	$\text{Li}_4\text{SiO}_4$	—	$\text{Li}_2\text{SiO}_3$	$\text{Li}_2\text{Si}_2\text{O}_5$	none
$\text{Na}^+$	1.02	$\text{Na}_4\text{SiO}_4$	—	$\text{Na}_2\text{SiO}_3$	$\text{Na}_2\text{Si}_2\text{O}_5$	none
$\text{K}^+$	0.75	$\text{K}_4\text{SiO}_4$	—	$\text{K}_2\text{SiO}_3$	$\text{K}_2\text{Si}_2\text{O}_5$	$\text{K}_2\text{Si}_4\text{O}_9$
$\text{Rb}^+$	0.67	$\text{Rb}_4\text{SiO}_4$	—	$\text{Rb}_2\text{SiO}_3$	$\text{Rb}_2\text{Si}_2\text{O}_5$	$\text{Rb}_2\text{Si}_4\text{O}_9$
$\text{Cs}^+$	0.61	$\text{Cs}_4\text{SiO}_4$	—	$\text{Cs}_2\text{SiO}_3$	$\text{Cs}_2\text{Si}_2\text{O}_5$	$\text{Cs}_2\text{Si}_4\text{O}_9$

the field strength around the cations, but so is the maximum degree of polymerization which silicates can have. This becomes apparent when the information compiled in Tables 1 and 2 is considered. In Table 1 some electropositive elements are arranged according to their position in the periodic table and underneath each element the most polymerized silicate known of this element is given. (Polymerization means in these cases sharing of O by two Si atoms. Throughout this paper only anhydrous silicates with but one kind of cation are considered.) The tendency to form silicates in general and polymerized silicates in particular increases from the upper right side of Table 1 toward its lower left side. The small highly charged ions like  $B^{3+}$  and  $C^{4+}$  ( $Ti^{4+}$ ) do certainly not form simple anhydrous silicates at all, whereas the large and low-charged alkali ions form silicates with a maximum degree of polymerization.

The elements in the table make noble gas type cations. In this case therefore, the ionic potential, charge/radius, is an adequate relative measure of the field strengths.<sup>1</sup> The ionic potentials of the cations are given in table 2 together with some of the monocationic anhydrous silicates which the elements are known to form. There is a very obvious correlation between ionic potential and the maximum degree of polymerization found in the silicates.

TABLE 3. HEATS OF FORMATION OF  $M_4SiO_4$  AND  $M_2SiO_4$  FROM COMPONENT OXIDES, GIVEN IN Kcal/MOL.<sup>1</sup>

Silicate	$-\Delta H_{298}$	Silicate	$-\Delta H_{298}$
$Li_4SiO_4$	$49.0 \pm 5$	$Be_2SiO_4$	$12.0 \pm 5$
$Na_4SiO_4$	$83.7 (?)$	$Mg_2SiO_4$	$15.1 \pm 1$
$K_4SiO_4$	$87.5 \pm 7$	$Ca_2SiO_4$	$32.8 \pm 0.5$
$Rb_4SiO_4$	—	$Sr_2SiO_4$	—
$Cs_4SiO_4$	—	$Ba_2SiO_4$	$46.0 \pm 7$

<sup>1</sup> Data from Kubaschewski and Evans (1951).

#### RELATIVE STABILITIES OF ORTHOSILICATES

Let us discuss the stabilities of orthosilicates relative to the free metallic oxide and silica. The known heats of formation show that the stability, measured in terms of heats of formation from oxides per two equivalent metal oxides,<sup>2</sup> increases with decreasing ionic potential (Table 3).

<sup>1</sup> If noble gas type ions are compared with non-noble gas type ions, the ionization potential would be a better indicator of relative field strengths than the ionic potential (See Ahrens (1953) and Ramberg (1953)).

<sup>2</sup> At low temperature the entropy term in the free-energy equation:  $\Delta F = \Delta H - \Delta ST$  is small for silicates so that the difference between heats of formation  $\Delta H$ , and free-energy of formation,  $\Delta F$ , is negligible.  $\Delta H$  is therefore a sufficient accurate stability indicator for our task.

This can now be explained qualitatively by means of a rather simple crystal-energetic model by considering the relative energies of the  $M-O$  and the  $Si-O$  bonds in the free oxides and in the orthosilicates respectively. ( $M^{z+}$  is the cation.) The  $Si-O$  bond is (usually) stronger in the orthosilicate ion than in quartz because the  $Si-O$  bond is more covalent, or  $O^{2-}$  is more polarized in  $SiO_4^{4-}$  than in quartz where oxygen is shared between strongly contrapolarizing  $Si^{4+}$  ions. On the other hand the  $M-O$  link is weaker in a silicate than in the metal oxide. The question whether or not an orthosilicate is stable relative to the free oxides depends then to a large extent upon the degree of strengthening of the  $Si-O$  bonds and weakening of the  $M-O$  bonds when metal oxides and quartz unite to form a silicate. The strengthening of the  $Si-O$  bond represents a decrease in energy thus tending to stabilize the silicate whereas the weakening of the  $M-O$  bond represents an increase in energy thus working against the formation of the silicate. The heat of formation of the silicate from the constituent oxides is then the difference between these two bond-energy changes.

When we now compare the energies of the  $Si-O$  and  $M-O$  bonds in the oxides and silicates respectively, we shall sacrifice some accuracy for the benefit of simplicity in order to make the basic principles clear. Thus, for example, repulsive energy between penetrating electron clouds will be disregarded. Furthermore the repulsive energy between oxygen ions and between the positive ions respectively, will be disregarded, hoping that this energy will be approximately equal in a silicate and in the corresponding free oxides. After these simplifications are made, the only energy difference to be expected between free oxides and equivalent amount of orthosilicate is that due to change of attractive energy of  $M-O$  and  $Si-O$  links in the oxides and the silicate respectively. Such an energy change is partly due to polarization of the oxygen ions, partly to covalent bonds. It is not easy to distinguish between polarization of  $O^{2-}$  and covalent sharing of electrons between oxygen and attached cation. Since the energy changes due to polarization can be calculated, at least in principle, whereas there is no easy way of calculating covalent bond energy in silicates, we shall treat the crystals from the viewpoint of varying degree of polarization of the oxygen ion. *It appears then that the stability of the silicates is closely related to the degree of polarization of  $O^{2-}$ , the more polarized it is the more stable are the silicates.*

In the simple metal oxides and in quartz each oxygen is more or less symmetrically surrounded by one kind of cation, the fields of which cancel out approximately in the center of the oxygen ion. One can therefore, as an approximation, consider the oxygen ions in the coordination lattices of the free oxides as not being polarized at all. The electrostatic energy of each  $Si^{4+}-O^{2-}$  link in quartz is therefore given by the simple equation:



$$(1) \quad E_q = -\frac{4e2e}{d_{\text{Si-O}}}, \text{ where } d_{\text{Si-O}} \text{ is the}$$

Si—O distance in quartz. The repulsive energy due to overlapping of the electron clouds is disregarded.

The electrostatic energy of each  $M^{z+}$ — $O^{2-}$  bond in the metal oxide is:

$$(2) \quad E_{M-O} = -\frac{ze2e}{d_{M-O}},$$

where  $d_{M-O}$  is the M—O distance, and any repulsive energy is disregarded. These electrostatic bond energies must now be compared with the energies of the corresponding bonds in the silicates where  $O^{2-}$  is polarized by the  $Si^{4+}$  ion. Each  $O^{2-}$  ion in the orthosilicates has one  $Si^{4+}$  and  $m$   $M^{z+}$  attached to itself. In  $Be_2SiO_4$ , for example, there are two  $Be^{2+}$  bound to each oxygen (i.e.,  $m=2$ ); in forsterite ( $Mg_2SiO_4$ ) there are three  $Mg^{2+}$  attached to each oxygen and the same is true for  $\gamma$   $Ca_2SiO_4$  and  $Ba_2SiO_4$  and possibly also for  $Sr_2SiO_4$  which all have the  $Al_2BeO_4$  type of structure (Wyckoff, 1951, vol. 2).

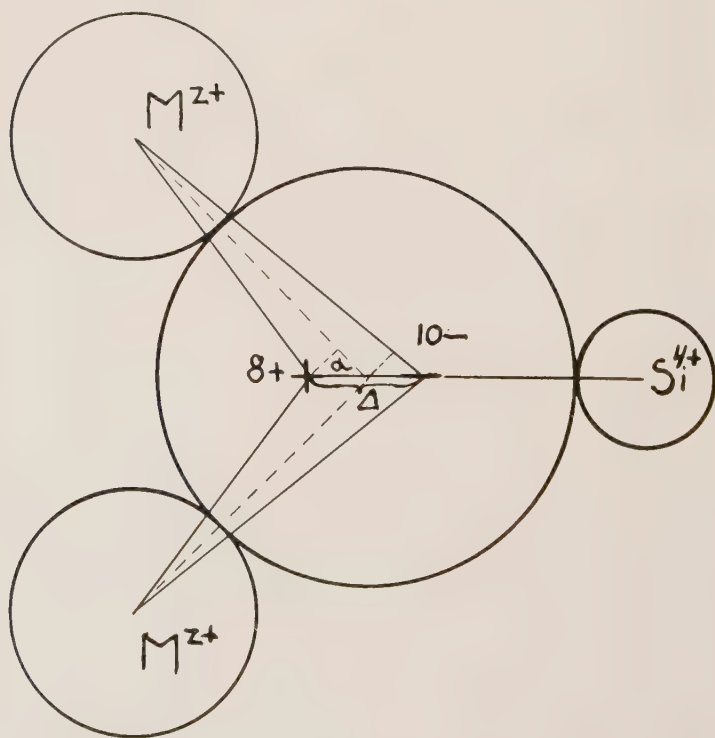


FIG. 1. Polarized oxygen ion surrounded by one  $Si^{4+}$  and two metal ions,  $M^{z+}$ .

In order to demonstrate the effect of polarization of  $O^{2-}$  we shall assume that  $Si^{4+}$  and the  $m M^{z+}$  ions are arranged in a symmetric fashion around  $O^{2-}$  in such a way that the resultant field of the  $m M^{z+}$  ions is parallel to the line through  $Si^{4+}$  and the center of oxygen (Fig. 1).  $O^{2-}$  is then situated in a field with a strength equal to the difference between the field caused by  $Si^{4+}$  and the resultant field of the  $m M^{z+}$  ions. For stable silicates the field around  $Si^{4+}$  is the stronger so that  $O^{2-}$  is unsymmetrically polarized, i.e. a dipole moment is induced in  $O^{2-}$  which may be pictured as if the 10 electrons on  $O^{2-}$  are displaced a distance  $1/2\Delta$  toward Si and the oxygen nucleus with its 8 positive charges is displaced  $1/2\Delta$  away from Si, (Fig. 1). For simplicity, let the distance from the center of the polarized  $O^{2-}$  to Si and M respectively, be  $d_{Si-O}$  and  $d_{M-O}$  as in the free oxides. The energy of  $Si^{4+}$  with respect to the polarized oxygen then is:

$$(3) \quad E'_{Si-O} = -\frac{4e10e}{d_{Si-O} - \frac{1}{2}\Delta} + \frac{4e8e}{d_{Si-O} + \frac{1}{2}\Delta}$$

where the first term represents the attractive energy between  $Si^{4+}$  and the 10 electrons on  $O^{2-}$ , and the second term represents the repulsive energy between  $Si^{4+}$  and the oxygen nucleus. Equation (3) can be rearranged to a more convenient form (see van Arkel, 1949, p. 143):

$$(4) \quad \begin{aligned} E'_{Si-O} &= -\frac{4e2e}{d_{Si-O} - \frac{1}{2}\Delta} - \frac{4e8e}{d_{Si-O} - \frac{1}{2}\Delta} + \frac{4e8e}{d_{Si-O} + \frac{1}{2}\Delta} \\ &= -\frac{4e2e}{d_{Si-O} - \frac{1}{2}\Delta} - \frac{\Delta 4e8e}{d_{Si-O}^2 - \frac{1}{4}\Delta^2} \end{aligned}$$

Since  $\Delta$  is usually very small relative to  $d_{Si-O}$ , the expression becomes:

$$(5) \quad E'_{Si-O} = -\frac{4e2e}{d_{Si-O}} - \frac{\Delta 4e8e}{d_{Si-O}^2}$$

where the first term is the same as the attractive energy between  $Si^{4+}$  and unpolarized  $O^{2-}$  in quartz, and the second term represents an additional attraction caused by the dipole induced in  $O^{2-}$  in the silicate.

With reference to the energy of a  $M^{z+}$  ion with respect to the polarized  $O^{2-}$  ion it is noted that the distance between  $M^{z+}$  and the center of gravity of the displaced electron cloud on  $O^{2-}$  is approximately  $d_{M-O} + 1/2\Delta \cos \alpha$ , where  $\alpha$  is the angle between the M—O joint and the induced dipole in  $O^{2-}$ , which in this symmetric and simple model is parallel to the Si—O joint. The distance between  $M^{z+}$  and the oxygen nucleus is approximately  $d_{M-O} - 1/2\Delta \cos \alpha$ . The electrostatic energy of  $M^{z+}$  with respect to the oxygen dipole is thus:

$$(6) \quad \begin{aligned} E'_{M-O} &= -\frac{ze10e}{d_{M-O} + \frac{1}{2}\Delta \cos \alpha} + \frac{ze8e}{d_{M-O} - \frac{1}{2}\Delta \cos \alpha} \\ &= -\frac{ze2e}{d_{M-O} + \frac{1}{2}\Delta \cos \alpha} + \frac{ze8e\Delta \cos \alpha}{d_{M-O}^2 - \frac{1}{4}\Delta^2 \cos^2 \alpha} \end{aligned}$$

or, if  $\Delta$  is very small relative to  $d_{M-O}$ :

$$(7) \quad E'_{M-O} = -\frac{ze2e}{d_{M-O}} + \frac{ze8e\Delta \cos \alpha}{d_{M-O}^2}.$$

The first term is the electrostatic energy of  $M^{z+}$  with respect to unpolarized  $O^{2-}$  and thus is identical to the energy of the  $M-O$  bonds in the metal oxide. The second term represents an additional energy of the  $M-O$  link due to the dipole moment of the polarized oxygen in the silicate.

When expression (5) is compared with expression (1) it is seen that the energy of a  $Si-O$  link in the orthosilicate anion is lower than the energy of this link in quartz, provided that the field around  $Si^{4+}$  is stronger than the resultant of the fields around the  $m$   $M^{z+}$  ions such that the electrons of  $O^{2-}$  are displaced toward  $Si^{4+}$ . Under the same conditions it is seen that the energy of the  $M-O$  link in the silicate (expression (7)) is higher than that of the same link in the metal oxide provided that  $\alpha < 90^\circ$  (so that  $\cos \alpha$  is positive). *It is noted that the decrease in energy of one  $Si-O$  bond is greater than the increase in energy of one  $M-O$  bond for cations which are larger and have lower charge than  $Si^{4+}$ .* If a silicate shall form from quartz and metal oxide exothermally, the decrease in energy due to changes in all the  $Si-O$  links in a crystal must be numerically larger than the increase in energy due to changes in all the  $M-O$  links in the same crystal. In order to consider this we must count  $Si-O$  and  $M-O$  links in an orthosilicate and in equivalent amounts of quartz and metal oxide. There are  $4N$   $Si-O$  links in an orthosilicate of type  $M_{4/z}SiO_4$  reckoned per mol  $Si$  atoms. ( $N$  is Avogadro's number.) In the same silicate there are  $4n/z$   $N$   $M-O$  links where  $n$  is the coordination number of  $M^{z+}$ , and  $z$  its charge. In an equivalent amount of  $SiO_2$  (one mol) there also are  $4N$   $Si-O$  bonds; and in an equivalent amount of metal oxide there are  $4n/z$   $N$   $M-O$  links provided that the coordination number of  $M^{z+}$  is the same as in the orthosilicate, which is actually true in many cases.

The electrostatic energy of all the  $M-O$  and  $Si-O$  links in the free oxides which are equivalent to one mol  $M_{4/z}SiO_4$  is then:

$$(8) \quad \sum E_{oxides} = 4NE_q + \frac{4}{z} nNE_{M-O} = -4N \frac{4e2e}{d_{Si-O}} - \frac{4}{z} nN \frac{ze2e}{d_{M-O}}$$

(see equations (1) and (2)). The energy of the corresponding bonds in the orthosilicate,  $M_{4/z}SiO_4$  is:

$$(9) \quad \begin{aligned} \sum E_{silicate} = 4NE'_{Si-O} + \frac{4}{z} nNE'_{M-O} = & -4N \left( \frac{4e2e}{d_{Si-O}} + \frac{4e8e\Delta}{d_{Si-O}^2} \right) \\ & - 4 \frac{n}{z} N \left( \frac{ze2e}{d_{M-O}} - \frac{ze8e\Delta \cos \alpha}{d_{M-O}^2} \right) \end{aligned}$$

where for simplicity all the  $M^{z+}$  ions which are linked to one  $O^{2-}$  ion are supposed to make the same angle,  $\alpha$ , with the dipole of the polarized oxygen which is parallel to the O—Si joint (Fig. 1).

The difference in electrostatic bond energy between the silicate and the free oxides is then:

$$(10) \quad \Delta E = \sum E_{\text{silicate}} - \sum E_{\text{oxides}} = 4N \left( -\frac{32e^2\Delta}{d_{\text{Si-O}}^2} + \frac{n8e^2\Delta \cos \alpha}{d_{\text{M-O}}^2} \right).$$

A part of this energy will be absorbed as internal energy of the polarized oxygen ions (See for example Slater, 1951, p. 394). This absorbed energy is about one half of  $\Delta E$ . Consequently the net energy change, or the heat of formation,  $\Delta H$ , of one mol  $M_{4/z}\text{SiO}_4$  from the free oxides becomes:

$$(11) \quad \Delta H = 16N\Delta e^2 \left( \frac{n \cos \alpha}{d_{\text{M-O}}^2} - \frac{4}{d_{\text{Si-O}}^2} \right).$$

$8e\Delta$  is the induced dipole moment of the oxygen ion. The dipole moment is proportional to the strength of the field in which oxygen is situated (see Van Arkel, 1949, p. 143 ff):

$$(12) \quad 8e\Delta = \bar{\alpha}F$$

where  $\bar{\alpha}$  is the polarizability of  $O^{2-}$  and  $F$  is the strength of the field. The strength of the field in which  $O^{2-}$  is situated is not constant but changes with distance from  $\text{Si}^{4+}$  and from the  $m$   $M^{z+}$  ions. For simplicity we shall consider the field in the center of oxygen as the average strength of the field which polarizes oxygen. This field is equal to the difference of the field created by  $\text{Si}^{4+}$  and the resultant field created by the  $m$   $M^{z+}$  ions. According to elementary electrostatics the strength of this field is:

$$(13) \quad F = \frac{4e}{d_{\text{Si-O}}^2} - \frac{mze \cos \alpha}{d_{\text{M-O}}^2}$$

if all the  $M^{z+}$  ions are situated in a symmetric fashion around the oxygen ion such that  $\alpha$  is constant (see Fig. 1). Hence:

$$(14) \quad 8e\Delta = \bar{\alpha} \left( \frac{4e}{d_{\text{Si-O}}^2} - \frac{mze \cos \alpha}{d_{\text{M-O}}^2} \right),$$

which, introduced into equation (12) gives:

$$(15) \quad \Delta H = 2Ne^2\bar{\alpha} \left( \frac{4}{d_{\text{Si-O}}^2} - \frac{mz \cos \alpha}{d_{\text{M-O}}^2} \right) \left( \frac{n \cos \alpha}{d_{\text{M-O}}^2} - \frac{4}{d_{\text{Si-O}}^2} \right).$$

Now the number  $m$ , of  $M^{z+}$  ions which are attached to each oxygen is related to the coordination number,  $n$ , of the cation and to its charge  $z$ . Assume that the number of cations which are attached to one  $O^{2-}$  ion is the same for all oxygen ions in a given orthosilicate and also that all  $M^{z+}$  in the silicate have the same coordination number with respect to oxygen. In this case one can reason as follows. There are four oxygens in

an orthosilicate of type  $M_{4/z}SiO_4$ , and there are  $4/z$   $M^{z+}$  ions each of which requires  $n$  oxygen ions in its coordination sphere. Each oxygen must then be shared among  $n/z$   $M^{z+}$  ions, hence:

$$(16) \quad m = \frac{n}{z}.$$

This value of  $m$  can be introduced in equation (15):

$$(17) \quad \begin{aligned} \Delta H &= 2Ne^2\bar{\alpha} \left( \frac{4}{d_{Si-O}^2} - \frac{n \cos \alpha}{d_{M-O}^2} \right) \left( \frac{n \cos \alpha}{d_{M-O}^2} - \frac{4}{d_{Si-O}^2} \right) \\ &= -2Ne^2\bar{\alpha} \left( \frac{4}{d_{Si-O}^2} - \frac{n \cos \alpha}{d_{M-O}^2} \right)^2. \end{aligned}$$

The quantities in these equations are constants except  $n$ ,  $\alpha$ , and  $d_{M-O}$ . It is perhaps surprising to find that the charge on the cation does not occur as a variable in the equation, only the radius of the cation (which is involved in the distance  $d_{M-O}$ ) and its coordination number with reference to oxygen, together with the bond-angle  $\alpha$ . The absence of  $z$  in equation (17) will be discussed later.

It is seen that if

$$\frac{n \cos \alpha}{d_{M-O}^2} \geq \frac{4}{d_{Si-O}^2}$$

the equation becomes negative, indicating that the silicate should be stable relative to the free oxides. The larger the difference between  $4/d_{Si-O}^2$  and  $n \cos \alpha/d_{M-O}^2$  the greater the numerical value of  $-\Delta H$  and hence the more stable the silicate.<sup>3</sup> If

$$\frac{n \cos \alpha}{d_{M-O}^2} = \frac{4}{d_{Si-O}^2}$$

$\Delta H$  becomes zero and the silicate is not stable except perhaps at  $0^\circ K$ . If

$$\frac{n \cos \alpha}{d_{M-O}^2} > \frac{4}{d_{Si-O}^2}$$

the resultant field of the  $M^{z+}$  ions is stronger than that of  $Si^{4+}$  such that the negative charge on  $O^{2-}$  is displaced toward the  $M^{z+}$  ions and the nucleus toward  $Si^{4+}$ . This situation is actually rare but if it occurred, a stable silicate might possibly result, because the energy change combined with the strengthening of the  $M-O$  bonds (relative to these in  $M$ -oxide) is numerically larger than the energy change combined with the weakening of the  $Si-O$  bond (relative to this in quartz). More commonly the condition exists:

<sup>3</sup> Note the previous remark on the relation between free-energy and total energy.



$$\frac{n \cos \alpha}{d_{\text{M-O}}^2} < \frac{4}{d_{\text{Si-O}}^2},$$

which means that the field around  $\text{Si}^{4+}$  is the stronger and oxygen has the negative end of its dipole closest to  $\text{Si}^{4+}$ . In this case the strengthening of the Si—O bond is more important than the weakening of the M—O bonds and energy will be released when the silicates form from the oxides. This is the usual situation found in stable silicates.

It is interesting to see how the values of  $\Delta H$  calculated from equation (17) compares with the empirically found stabilities of some orthosilicates as for example  $\text{Be}_2\text{SiO}_4$ ;  $\text{Mg}_2\text{SiO}_4$ ;  $\gamma$   $\text{Ca}_2\text{SiO}_4$ ;  $\text{Sr}_2\text{SiO}_4$  and  $\text{Ba}_2\text{SiO}_4$ . In  $\text{Be}_2\text{SiO}_4$ ,  $\text{Be}^{2+}$  is surrounded by 4 oxygen ( $n=4$ ), in the other cases the cations are surrounded by 6 oxygen ( $n=6$ ). For simplicity we shall assume that the angle  $\alpha$  is the same for all these silicates so that all cations about an  $\text{O}^{2-}$  ion make the same angle with the dipole of the polarized oxygen. Let this angle for example be  $60^\circ$ . The Si—O distance is taken as  $1.65\text{\AA}$  whereas the M—O distances are considered to be the sum of the ionic radii.

Table 4 shows some of the terms of equation (17) based upon these premises together with the *relative*  $\Delta H$ -values computed from equation (17). *It is seen that the trend of the calculated  $\Delta H$  values is the same as for*

TABLE 4. CALCULATED AND MEASURED HEATS OF FORMATIONS  
OF ORTHOSILICATES

Silicate	Approximate cation-oxygen distances (A)	Coordination number $n$	$\left( \frac{4}{d_{\text{Si-O}}^2} - \frac{n \cos \alpha}{d_{\text{M-O}}^2} \right)$	Relative $\Delta H$ calculated, eq. (17) (common factor: $2\text{Ne}^2\bar{\alpha}$ )	$\Delta H$ measured, kcal per mol. $\text{M}_2\text{SiO}_4$
$\text{Be}_2\text{SiO}_4$	1.66	4	$1.47 - \frac{2}{2.76}$	— .41	$-12 \pm 5$
$\text{Mg}_2\text{SiO}_4$	2.10	6	$1.47 - \frac{3}{4.41}$	— .62	$-15 \pm 1$
$\text{Ca}_2\text{SiO}_4$	2.38	6	$1.47 - \frac{3}{5.66}$	— .70	$-32.8 \pm 0.5$
$\text{Sr}_2\text{SiO}_4$	2.59	6	$1.47 - \frac{3}{6.71}$	— 1.04	—
$\text{Ba}_2\text{SiO}_4$	2.75	6	$1.47 - \frac{3}{7.56}$	— 1.14	$-46.0 \pm 7$

the actually measured heats of formation. It should be emphasized, however, that the crystal model employed above is too simple to allow correct absolute  $\Delta H$  values to be calculated for the various silicates. The model does lead to calculated relative heats of formation of orthosilicates that are in agreement with experimental calorimetric data.<sup>4</sup>

The fact that the charge on  $M^{z+}$  does not occur in equation (17) should be discussed. This is a result of the crystal model employed being too simple. However, it can be modified to account qualitatively for change of cationic charge. It is evident from geochemical observation, as well as from heat-of-formation measurements, that the charges on the cations, not only their radii and coordination number, influences the stability of the silicates (see table (1) and (2)). The reason for the charge on  $M^{z+}$  not appearing in equation (17) is essentially due to the simple picture which was employed regarding the polarization of  $O^{2-}$ . It was assumed that the electron cloud around the oxygen nucleus was a rigid construction which during polarization was displaced relative to the nucleus so that the center of gravity of the electron cloud in the polarized oxygen fell outside the nucleus thus inducing a dipole moment in  $O^{2-}$ . In this model the relative displacement of nucleus and electron cloud of  $O^{2-}$  (situated between one  $Si^{4+}$  and  $m$   $M^{z+}$  in symmetric configuration) is given by the field of  $Si^{4+}$  minus the resultant field of the  $m$   $M^{z+}$  ions. In other words this model will not respond to variable field strengths around the individual  $M^{z+}$  ions, but only to the resultant field from all of them (measured in the center of  $O^{2-}$ ). It can be shown that this resultant field will be independent of the charge on each  $M^{z+}$  (as long as  $n$  and  $\alpha$  are the same) because if the charge increases, the number of cations around each  $O^{2-}$  decreases and *vice versa*. In an orthosilicate of type  $M_{4/z}SiO_4$  the number of cations,  $m$ , which is attached to each oxygen is determined by the charge,  $z$ , and the coordination number,  $n$ , of the cation with reference to  $O^{2-}$  by the following relation:  $m = n/z$ , as discussed previously. Each cation has  $z$  positive charges and the total number of positive charges carried by the  $m$  cations is:  $z \cdot m = z \cdot n/z = n$  which shows that the total number of charges carried by the  $m$  cations of charge  $z$  is simply equal to the coordination number of  $M^{z+}$  relative to oxygen. Suppose, for example, that in  $Li_4SiO_4$ ,  $Mg_2SiO_4$  and in a hypothetical compound  $TiSiO_4$  the cations all have coordination number 6. Then there must be 6  $Li^+$ , 3  $Mg^{2+}$  and 1.5  $Ti^{4+}$  ions respectively attached to each oxygen. (This could be true for every oxygen in the crystals, but the figures given can also rep-

<sup>4</sup> It should be remembered in this connection that the polarizability of an anion in a crystalline structure is not the same as its polarizability as a free ion. The polarizabilities of anions in crystals are not known, and equation (17) cannot therefore be used to calculate quantitative energy relations.

represent the average number of cations attached to each oxygen. This must evidently be the case for a hypothetical compound  $\text{TiSiO}_4$ —in which some oxygens may have one  $\text{Ti}^{4+}$  attached, others two  $\text{Ti}^{4+}$  if the coordination number of all  $\text{Ti}^{4+}$  is 6). This means that the number of positive charges carried by 6  $\text{Li}^+$ , 3  $\text{Mg}^{2+}$  and 1.5  $\text{Ti}^{4+}$  is 6 in all cases. These charges are approximately at the same distance from the center of  $\text{O}^{2-}$  because the radii of  $\text{Li}^+$ ,  $\text{Mg}^{2+}$  and  $\text{Ti}^{4+}$  are very nearly equal. Let also the angle  $\alpha$  be constant. It follows then that they give the same resultant field in the center of oxygen and consequently that the 6  $\text{Li}^+$ , the 3  $\text{Mg}^{2+}$  and the 1.5  $\text{Ti}^{4+}$  ions polarize  $\text{O}^{2-}$  to exactly the same extent, provided that the electron cloud around  $\text{O}^{2-}$  is displaced as a rigid body relative to the nucleus as was assumed in the model above.

If the picture of the polarized oxygen is made less simple but more in harmony with what really happens, it appears that the charge on  $\text{M}^{z+}$  is of great significance. The electron cloud around oxygen is a rather flexible construction which does not only move as a whole in response to electrostatic fields but within which adjustments take place locally in response to the fields of each individual cation attached. In other words, the electron density close to each cation is higher than normal. This means additional attraction which is going to make a configuration like  $\text{Li}_4\text{OSi}$  different in respect to energy than, for example,  $\text{Mg}_2\text{OSi}$ . It is sufficient for our qualitative model to assume that the increment of electron density (above normal) in the electron cloud of  $\text{O}^{2-}$  close to a cation,  $\text{M}^{z+}$ , is proportional to the field around the cation:

$$(18) \quad \eta = \frac{ze}{r^2} \mu$$

where  $\eta$  is the excess electron density in a volume with mean distance  $r$  from the cation.  $\mu$  is a proportionality factor depending on the deformability of the electron cloud. ( $\mu$  is not the same as the polarizability,  $\bar{\alpha}$ , of  $\text{O}^{2-}$  as a whole).  $z$  is the charge on the cation. The additional attraction between  $\text{M}^{z+}$  and the extra concentration of electrons close to the ion is then

$$(19) \quad -\frac{ze\mu}{r} = -\frac{z^2e^2\mu}{r^3}$$

if the repulsive energy due to overlapping of the electrons from  $\text{O}^{2-}$  and  $\text{M}^{z+}$  is disregarded. Now the deformability,  $\mu$ , of the electron cloud of  $\text{O}^{2-}$  is not the same in a metal oxide and in a silicate where the strongly polarizing  $\text{Si}^{4+}$  ion pulls the electrons of  $\text{O}^{2-}$  toward the  $\text{Si—O}$  link.  $\mu$  for the oxygen in most metal oxides should be somewhat larger than for oxygen bound to silicon in orthosilicates because most metal ions have less polarizing power than  $\text{Si}^{4+}$ . The difference in energy due to the kind of

polarization considered above, of a M—O link in metal oxide and in silicate is:

$$(20) \quad \frac{z^2 e^2}{r^3} (\mu_{ox} - \mu_{sil})$$

where  $\mu_{ox}$  is the deformability of the oxygen electron cloud in the metal oxide and  $\mu_{sil}$  the deformability of the oxygen electron cloud in silicate. In most cases (when  $M^{z+}$  has less polarizing power than  $Si^{4+}$ )  $\mu_{sil} < \mu_{ox}$  showing that the part of the M—O bond which is due to this kind of polarization is stronger in the oxide than in corresponding silicate. In other words *the free oxides are more stable relative to the silicates than equation (17) indicates*. It is noted that the energy difference is proportional to the square of the charge on  $M^{z+}$ . When the term above (eg. (20)) is added to equation (17) it is seen that the heat of formation of orthosilicates from the component oxides is also influenced by the charge of the cation in such a fashion that the larger the charge the less stable the silicate relative to the oxides. In other words  $\Delta H$  will increase (assume smaller negative values or even be positive) with increasing  $z$ . This may be shown by an equation like the following:

$$(21) \quad \Delta H = 2N\bar{\alpha}e^2 \left( \frac{4}{d_{Si-O}^2} - \frac{n \cos \alpha}{d_{M-O}^2} \right) \left( \frac{n \cos \alpha}{d_{M-O}^2} - \frac{4}{d_{Si-O}^2} \right) + \frac{4}{z} nN \frac{z^2 e^2}{r^3} (\mu_{ox} - \mu_{sil})$$

where the notations are as defined above. ( $4/zn$  N is the total number of M—O links in one mol  $M_{4/z}SiO_4$ ).

This last modification of the crystal model is then in qualitative harmony with the observed facts concerning the effect of ionic charge on the relative stabilities of the various orthosilicates. The model is certainly still too simple to permit quantitative calculations of heats of formations because factors like geometric strain in the lattices, the effect of repulsive forces on the polarization and the participation of covalent binding are neglected. On the other hand the silicate model described above explains in a simple manner the major trend of the relative stabilities of silicates of various elements.

In the following we shall see that the repulsive forces may be essential for the discussion of some stability relations of polymerized silicates.

#### STABILITIES OF POLYMERIZED SILICATES

It is worth noting that the maximum degree of polymerization of the silicate anion is related to type of cation (Table 2). Large and weakly charged cations are much more apt to form polymerized silicates than small and highly charged cations (only anhydrous silicates with but one kind of cation is still considered). This observation should be explainable

by the crystal energy model discussed above if it is essentially correct. The fact that polymerized silicates are stable at all can be related to a varying polarizing power of the  $\text{Si}^{4+}$  ion. If silicon is surrounded by four unshared oxygens in an orthosilicate anion the polarizing power of  $\text{Si}^{4+}$  is relatively low because the electrons from all four surrounding  $\text{O}^{2-}$  ions are pulled rather close to  $\text{Si}^{4+}$  thus partly neutralizing its positive charge. In a  $\text{SiO}_4$  tetrahedron where the silicon ion is surrounded by shared oxygens also, the electrons on the shared  $\text{O}^{2-}$  ions in the  $\text{Si}-\text{O}-\text{Si}$  bridge are not pulled very close to the  $\text{Si}^{4+}$  ion because of contrapolarization by the opposite  $\text{Si}^{4+}$  ion. *In consequence the effective polarizing power of  $\text{Si}^{4+}$  increases with increasing number of shared oxygen ions such that the polarization of each unshared oxygen by  $\text{Si}^{4+}$  is stronger the more shared oxygens that remain around the silicon ion.* The energy released by breaking a  $\text{Si}-\text{O}-\text{Si}$  bridge and making two unshared  $\text{O}-\text{Si}$  bonds, therefore, increases with increasing number of shared  $\text{Si}-\text{O}-\text{Si}$  bridges which remain around the Si ions in the polymerized silicate anions. When an orthosilicate anion forms, the energy release is low, when a metasilicate anion forms, somewhat more energy is released, and when  $\text{Si}_2\text{O}_6^{2-}$ -type anion forms still greater energy is released. This is clearly seen when the heat of formation from oxides of silicates with various degrees of polymerization are considered (Table 5). In this Table the heat of formation is given per a constant (two) equivalent of metal oxides in order to insure that the same number of  $\text{Si}-\text{O}-\text{Si}$  bridges are broken in all cases.

TABLE 5. HEATS OF FORMATION OF VARIOUS POTASSIUM SILICATES FROM OXIDES, GIVEN IN kcal PER TWO EQUIVALENTS<sup>1</sup>

Silicate	$\frac{1}{2}\text{K}_4\text{SiO}_4$	$\text{K}_2\text{SiO}_3$	$\text{K}_2\text{Si}_2\text{O}_6$	$\text{K}_2\text{Si}_4\text{O}_9$
$-\Delta H_{298}$	$43.8 \pm 3$	$64.5 \pm 7$	$74.5 \pm 7$	$81.0 \pm 7$

<sup>1</sup> Data from Kubaschewski and Evans (1951).

The decreasing stability of polymerized silicates (decreasing numerical value of  $\Delta H$ ) with decreasing radius and increasing charge on the cation is explainable by the same arguments as were used for the relative stabilities of the orthosilicates. However, the polymerized silicates offer a somewhat different problem: why do some of the smaller and high-charged cations form orthosilicates (or pyrosilicates) but not meta- or  $\text{Si}_2\text{O}_6^{2-}$ -type silicates? And why do ions which are somewhat larger and/or have less charge form ortho- and metasilicates but not  $\text{Si}_2\text{O}_6^{2-}$ -type silicates, leaving the most highly polymerized silicates to the largest or lowest charged cations? This is a more difficult problem and an expla-



nation can only be suggested. The energy released when a metal oxide and quartz formed a silicate was considered essentially in two terms (1) the increased strength of the Si—O bond which releases energy, and (2) the decreased strength of the M—O bond which absorbs energy. For stable silicates the former is somewhat larger than the latter thus giving rise to exothermic formation of the silicate. Now in polymerized silicates the electron cloud of the unshared oxygen ion is pulled closer in toward  $\text{Si}^{4+}$  than in less polymerized or unpolymerized silicates, this being the chief reason why more heat is liberated when a stable polymerized silicate forms than when an orthosilicate forms with same cation. The closer to the  $\text{Si}^{4+}$  ion the electrons of  $\text{O}^{2-}$  are pulled, however, the stronger the repulsive force acting on these electrons because they penetrate deeper into the electron cloud of  $\text{Si}^{4+}$ . Hence the energy released *per distance* which the electrons on  $\text{O}^{2-}$  are pulled in toward the  $\text{Si}^{4+}$  ion is smaller and smaller the closer the electrons are pulled up to the  $\text{Si}^{4+}$  ion. The increase of energy of a M—O link *per distance* which the electrons on  $\text{O}^{2-}$  are pulled *away* from  $\text{M}^{z+}$  is proportional to the charge on  $\text{M}^{z+}$  and inversely proportional to the M—O distance. It seems possible then that if the metal cation ( $\text{M}^{z+}$ ) has a relatively strong field the energy increase of the M—O bond which is connected with increasing polymerization in the silicates, is larger than the energy decrease of the Si—O bond. This would offer a qualitative explanation why orthosilicates of ions with large charge or small radii are stable but not highly polymerized silicates of such ions.

With reference to the low stability of heavy metal silicates it can readily be explained by the model presented in this paper when one remembers that these metals make ions with much stronger fields than what should be expected on the basis of their radii. Such metals ( $\text{Zn}^{2+}$ ,  $\text{Cu}^{2+}$ ,  $\text{Ni}^{2+}$ ,  $\text{Co}^{2+}$ ,  $\text{Fe}^{2+}$ ,  $\text{Fe}^{3+}$ ,  $\text{Pb}^{2+}$ , etc.) behave somewhat similarly to noble gas type ions with small radius or high charge.

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## MEMORIAL OF A. ALBERT KLEIN

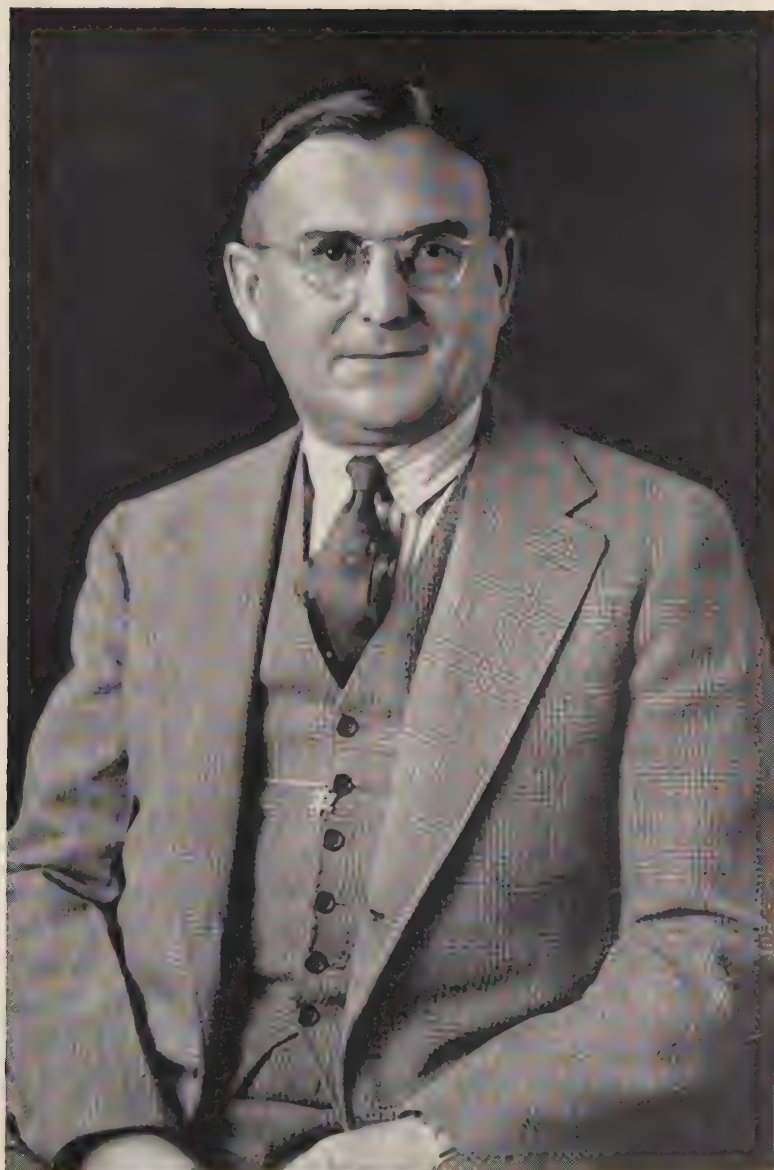
NEWMAN W. THIBAUT, *Norton Company, Worcester, Massachusetts.*

The sudden and wholly unexpected passing of A. Albert Klein on the evening of August 25, 1953, was a shock which will long be felt not only by his host of friends, but by the entire community as well. Unusually young in appearance, action and attitude, "Al" had left the laboratory in his usual good spirit, but suffered a fatal heart seizure as he was relaxing at home after the evening meal.

He was born in Syracuse, New York, May 6, 1889, oldest son of Ella (Kogan) and Samuel David Klein. He attended the local public schools, and graduated from high school in 1905, valedictorian of his class. During the next two years he was employed in the laboratory of the Semet-Solvay plant near Syracuse. Then with the encouragement and counsel of Dr. Edward H. Kraus, who had been his chemistry teacher in the Syracuse High School, he enrolled in the University of Michigan as an engineering student in the fall of 1907. Shortly afterwards, however, he transferred to Liberal Arts, specializing in mineralogy and received his B.S. degree in 1911 under Professor Kraus, who at that time was Director of the Mineralogical Laboratory. "Al" paid his own way through Michigan by assisting in the laboratories his last two years, and playing the violin in college orchestras. He was ever appreciative of the inspiration and assistance given him during his college days by Professor and Mrs. Kraus of whom he spoke with great affection on many occasions during the seventeen years of my association with him. There is no doubt that Professor Kraus contributed more to "Al's" personality than anyone else outside of his immediate family during this formative period of his life.

After graduation, he accepted the position of Assistant Physicist at the National Bureau of Standards, Washington, D. C., remaining in that capacity there and in Pittsburgh, Pennsylvania, from 1911 to 1916. During that period he applied his proficiency in optical mineralogy and physical chemistry to petrographic studies of porcelain, silica brick and Portland cement. With associates at the Bureau, he published a number of papers on these subjects as indicated in the appended bibliography. Those on Portland cement which were issued as *Technologic Papers of the Bureau of Standards* were monumental works of the day, eliciting requests for reprints nearly forty years later.

When he joined the staff of the Research Laboratories of Norton Company, Worcester, Massachusetts in 1916, he was one of the first, if



A. ALBERT KLEIN  
1889-1953

not the very first, petrographer employed full-time in American industry. With this Company he spent 37 years, first as Research Engineer and for the last twelve years as Assistant Director of Research and later of Research and Development, applying mineralogical techniques, particularly petrography, toward the solution of problems in the abrasive and refractory fields.

So successful was he in these endeavors that he was constantly consulted by personnel from every segment of the Norton organization. He published but few papers, but in addition to thousands of private reports, he contributed immeasurably to the Company and the entire abrasive industry through the advice and suggestions which he freely gave, without thought of personal credit, to all who consulted him. It was particularly significant that many of the younger men sought his counsel on matters often far removed from his chosen field.

"Al's" main interest was always ceramic petrography. Through years of constant contact he had developed that sixth sense which often permitted seemingly intuitive identification of compounds almost at a glance—a marvel to those yet uninitiated. At first he was somewhat reserved in accepting *x*-ray diffraction, but quickly appreciated its value once equipment had been installed. However, he always rightly insisted that it complement rather than replace examination with petrographic or metallographic microscope. Although in direct charge of the microscopic, physical research and chemical sections of the Norton Worcester laboratories, he personally made most of the petrographic examinations to the day of his death.

"Al" and I rarely disagreed as to observation or identification, but discussion was sometimes spirited, occasionally seemingly poignant, with respect to interpretation, consequence or prospects. Through it all, however, there was never a bit of animosity; invariably he closed the matter uproariously with an anecdote appropriate to the occasion.

In partial recognition of his accomplishments he was elected to fellowship in the Mineralogical Society of America, the American Ceramic Society and the American Association for the Advancement of Science. He was a member of the American Crystallographic Association, the American Society for Testing Materials where he was chairman of one sub-committee and member of another, the Worcester Engineering Society and Sigma Xi. He was chairman of the Research Technical Sub-Committee of the Grinding Wheel Institute.

Indefatigable in pursuit of his chosen field during working hours, the remainder of his time was occupied by numerous religious, cultural and civic interests which he pursued with his wife Ann (Shapero) whom he had known since childhood, and to whom he was married on December 28, 1913.



In 1921 they pioneered the establishment of Liberal Judaism in the city of Worcester; they nurtured the foundling with such zeal and devotion that today the outward embodiment, Temple Emanuel, is one of the great synagogues of the nation. Albert Klein drafted the constitution and by-laws, was chairman of the religious committee, director of the choir and for the last thirteen years was president of the Temple. Because of his scientific training he was unusually successful in delving to the core of a problem, seeking facts with utmost fairness, thereby resolving divergent opinions into united action for the common good. In this work, likewise, he gave unstintingly of his time and financial means without thought of public acclaim, preferring to remain merely as another member of the large congregation. He was also active in Jewish cultural and charitable organizations and the establishment of a Jewish homeland in Israel.

But his outside activities were by no means limited by his faith; rather they transcended creed, color and all barriers to the brotherhood of man. He was active in such organizations as the Community Chest, Boy Scouts, Masons and Community Council. During World War II he was a member of one of the local Selective Service Boards which occupied much of his time for several years. He was particularly conscientious, and insisted that all cases be carefully investigated from every point of view in fairness both to the boy and the nation.

He was an accomplished musician, and at one time considered becoming a professional violinist. In his college days he was manager of the band. Later he was a member of local symphony orchestras, but became less active as a player in recent years, devoting most of his musical talent to affairs at the Temple.

Two children were born to Ann and Albert Klein. One, David Louis, is President of Pul-Ver-Ite, Inc., Brooklyn, New York, the other, Dr. Donald Charles, is Executive Director of the Human Relations Council, Inc., at Wellesley Hills, Massachusetts. There are five grandchildren.

The loss of "Al" Klein will long be felt by his family, his associates at Norton Company, the Grinding Wheel Institute and at Temple Emanuel, by his many friends and acquaintances in all walks of life, and by the whole community itself.

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## MEMORIAL OF OLAN IVAN LEE

HERMAN YAGODA, *National Institutes of Health, Bethesda, Maryland.*

Olan Ivan Lee, Chief of the spectrographic and microchemical sections of the United States Testing Co., died at the Christ Hospital, Jersey City, New Jersey, on the 26th of November, 1952. Although most of his professional activities were in the fields of chemistry and spectroscopy, he will be best remembered by his numerous friends for his profound interest and contributions to mineralogy.

If one were to attempt to bring to a focus the outstanding characteristics of an individual, then within the limits of terse phraseology, the personality of Ivan Lee crystallizes as the scholar in industry. He possessed the genial quality of introducing the academic approach in the mundane industrial laboratory, and of infecting associates with his enthusiasm for mineralogy, rare books on the physical sciences and the history of gemology. His home was a veritable museum of fine mineral specimens, largely collected during field trips while on brief vacations from the laboratory, and formed an enchanting environment for the entertainment of his friends. An evening spent with Ivan Lee and his wife, Mary, was not only pleasant in the usual amenities of life, but was an intellectual stimulus. The rare specimen of samarskite in his hand ceased to be a black amorphous mass dug up from a long-abandoned mine, but in the narration became a storehouse of rare elements with details of how best to isolate them, and a mystery whose composition was to be resolved by spectroscopy and neat microchemical tests. In the early hours of the morning, amidst the aroma of coffee, one learned that former President Hoover was the erudite translator of Agricola's "*De Re Metallica*," and that certain varieties of tectite were probably chips from the surface of the moon.

Ivan Lee was born on April 17, 1888, at Madison, Lake County, Ohio. His family moved East several years later, and his early youth was spent largely in New York City. He graduated from Brooklyn Polytechnic Institute in 1911 with a B. Sc. degree in chemistry. He interspersed his industrial work with postgraduate studies at Columbia University and special courses in microscopy at the Rochester plant of Bausch and Lomb. A mastery of spectroscopy led to numerous researches on the minor components of minerals and ores, and also provided a basis for many analytical procedures for trace constituents during his tenure as principal spectrographer for the Crocker Cancer Institute of Columbia University.

Ivan Lee was an active member of the mineralogical societies in the Metropolitan area and gave many lectures on his field trips and on re-



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cent advances in mineralogy. He was one of the founders of the New Jersey Mineralogical Society and served as president of the New York Mineralogical Club (1945–1947). He was one of the early members of the Mineralogical Society of America and was elected a fellow in 1923.

During his active life he found opportunity to publish some thirty-four papers in chemistry, engineering, mineralogy and spectrography. Of outstanding interest to mineralogists is his discovery of the reversible photosensitivity of hackmanite, and a review article on the mineralogy of hafnium prepared about three years after the discovery of the element. A select bibliography of his publications in the field of mineralogy is appended.

Olan Ivan Lee is survived by his wife, Mary Stewart, and a daughter, Virginia Lee.

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## MEMORIAL OF PAUL NIGGLI

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With the death on January 13, 1953, of Paul Niggli, since 1920 Professor of mineralogy and petrography in the Swiss Federal Institute of Technology at Zürich, the Earth Sciences have lost one of their most notable exponents and his co-workers an inspiring leader.

The main biographical details of his life are simple. Born on June 26, 1888, in the little Swiss town of Zofingen, he made preliminary contacts with the science that was later to become his domain while still a boy at school at Aarau. Coached by Professor Mühlberg, a well-known Swiss geologist who was his science teacher, he started to study and map the geological features of his native canton. By 1907, when he matriculated as a student at the Institute of Technology in Zürich, these investigations had furnished him with the material for a report that was published in due course by the Geological Commission of Switzerland. To the experience and pleasure in field work thus gained in his early days may be ascribed the value attached by him to observations in situ and the mastery in the art of unfolding to the participants in his field trips the subtle implications of the geological scene before them. Though the Geological Institute in Zürich was at the time of his entry in the hands of Professor Albert Heim, it was not this eminent teacher and scientist who modelled Niggli's ultimate career. For at that stage of his development fossils, sedimentary rocks, and tectonics offered Niggli too little scope for the application of physics and chemistry to be wholly satisfying objects of study. Minerals, igneous rocks and metamorphics which were included in the syllabus of the adjoining Institute under Professor Ulrich Grubenmann presented, on the contrary, a field entirely to his taste. It was thus in the main as a pupil of Grubenmann that Niggli presented himself in 1912 for the Ph.D. examination at the University of Zürich. His study on the chloritoid-bearing schists of the north-eastern Gotthard Massif submitted to the faculty as his dissertation reveals the interesting fact that many problems that were to remain life-long pre-occupations were already foremost in his mind. After a year spent in the Geophysical Institution of Washington, Niggli returned to Zürich and became lecturer (*Privatdozent*) in Zürich University. In 1914 he was, however, called to the University of Leipzig as extraordinary, and thereafter (1918) to Tübingen as ordinary Professor of Mineralogy. When in 1920 Professor Grubenmann resigned from the chair of mineralogy and petrography in Zürich, Niggli was elected to be his successor and in spite



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of calls that came to him from abroad, did not again leave the Institute at which he himself had studied.

To call Niggli a mineralogist, petrographer, or geochemist, would be doing him less than justice. He was, in fact, all these things in a measure rarely attained by a single individual. The work carried out by him during his thirty-three years at Zürich can only be described as amazing, and a glance at the list of his scientific publications suffices to prove the aptness of this adjective. For the bibliography shows that during this time he published over 15, mostly voluminous, books as well as some two to three hundred papers, essays, etc., in various periodicals. These works

are in the main devoted to matters of petrography, mineralogy, crystallography, and crystal structure, but also include such bearing on questions of general scientific principles and methods, education, politics, history, philosophy, etc. It must be remembered, however, that only a limited part of his time could be devoted to scientific work. For beside a very full programme of lectures during term time and the hours devoted to his students, an interminable series of examinations as well as much administrative work both within the Institute and in the innumerable commissions and committees to which he belonged remained to be dealt with. He was for some years (1921 to 1940) editor of the "*Zeitschrift für Kristallographie*," but resigned from that post when political conditions in Germany took a turn that he disliked intensely.

Niggli had the satisfaction of receiving a very full measure of appreciation for his labours. He was awarded honorary academic degrees by the Institutes of Technology at Stuttgart and Karlsruhe and by the Universities of Geneva, Budapest, Sofia, and Liège. From America honours came to him from the Mineralogical Society (Correspondent in 1931 and Roebling Medallist 1947), the Geological Society (Correspondent 1935), the N. Y. Academy of Sciences (honorary life-member 1944), and the Academy of Natural Sciences of Philadelphia (Hayden Memorial Geological Award 1947). Beside these distinctions he was correspondent, honorary member, foreign member, etc., of learned societies in Austria, Belgium (3), Brazil, Finland (2), France (2), Germany (5), Great Britain (3), Holland, India (2), Italy (4), Poland, Roumania, Spain, Sweden, and U.R.S.S., and of half a dozen or more Societies in his own country.

For those who were closely associated with Niggli and have witnessed the gradual building up of his life's work, it is interesting to compare his early publications with those of later years. Two books published before coming to Zürich come clearly from the *investigator* Niggli. Thus his treatise on the volatile constituents in the magma (1918) is an enquiry into the physical chemistry of magmatic systems and reveals him as a master of his subject. The "Geometrische Kristallographie des Diskontinuums" (1919), too, was Niggli's fundamental contribution to a branch of science then still in its infancy. His volume (written with P. J. Beger) on rock and mineral provinces (1923) which contains the first catalogue of magma types based on molecular (so-called "Niggli") values and the treatise on rock metamorphism (1924) with Grubenmann as co-author are further products of a period during which he was actively concerned with practical research. If it was the *teacher*, Niggli who was largely responsible for many of the later works, this was, perhaps, due to the great success that attended his "Lehrbuch der Mineralogie" published in 1920. The freshness and originality of his treatment of the fundamental aspects

of crystallography, crystal chemistry, and crystal physics (with which was coupled a rather detailed account of the occurrence and deposits of minerals and rocks in the earth's crust) constituted a complete break-away from the general trend of German textbooks at that time and was very enthusiastically received by readers of widely different status. It henceforth became his ambition to keep this work abreast of the rapid development of the science and the repeated calls for new editions were responded to by several complete recastings and amplifications of the work, involving an ever broadening scope in the matter treated. The didactic approach to his subject can clearly be discerned in much of his later writing and led to his last great publication, the "Gesteine und Minerallagerstätten," the third volume of which remained unfinished at the time of his death.

Niggli's treatment of the "Earth Sciences" was conceived on a generous, indeed monumental, scale. Not only did he include in it every ingredient of the crust from igneous rocks to snow crystals, but all their manifestations as well, from the atomic assemblage to world distribution. His genius is attested by the fact that the universality of his interests was matched by the versatility of his ideas.

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Dr. Niggli's bibliography is so extensive that only a summary under the various headings will be recorded here. A complete list of publications has been prepared by *J. Marquard* and *J. Schroeter* and will be found in *Schweiz. Min. Petr. Mitt.*, Band 33, pp. 9-20, 1953.

Kristallstruktur- und Symmetriellehre: 3 books; 61 papers.

Allgemeine und Spezielle Mineralogie: 7 books; 25 papers.

Minerallagerstätten: 1 book; 14 papers.

Allgemeine Petrographie und ihre Physikalisch-Chemischen Grundlagen: 7 books; 63 papers.

Regionale Petrographie und Geologie: 4 books; 30 papers.

Beziehungen von Mineralogie und Petrographie zu Praktischen Fragen: 19 papers.

Schneeforschung: 4 papers.

Allgemeines zu den Mineralogischen Wissenschaften: 1 book; 12 papers.

Allgemeine Forschungs- und Unterrichtsfragen der Wissenschaften: 1 book; 24 papers.

Biographische Artikel: 5 papers.



## MEMORIAL OF FREDERICK EUGENE WRIGHT

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Fred(erick) E(ugene) Wright, a fellow and a past-president of the Mineralogical Society of America, passed away at his summer home on Sagastaweka Island, in the Thousand Islands near Gananoque, Ontario, on August 25, 1953. He will be missed by his many friends in mineralogy, geology, optics, astronomy, and in the army. His has been a brilliant scientific career with wide interests and many accomplishments.

Dr. Wright was born at Marquette, Michigan, October 16, 1877, where his father was stationed as State Geologist. The elder Wright had received training in mineralogy and geology in Germany and Sweden (1869-71) and was one of the first in this country to prepare thin sections of rocks. He made extensive contributions to the Pre-Cambrian geology of the Lake Superior region. The elder Wright died at an early age in 1888, and shortly after his death Mrs. Wright with her three sons moved to Ann Arbor, Michigan, where Fred Wright attended the public schools and was graduated from Ann Arbor High School in 1895.

The family then moved to Germany where Fred, the oldest of the three boys, was a student at the Realgymnasium at Weimar for one year. In 1896 he enrolled at the University of Heidelberg, where, following in the footsteps of his father, he undertook intensive studies in mineralogy, petrology, and geology, as well as in chemistry, physics, and mathematics, under the direction of such distinguished scientists as Harry Rosenbusch, Victor Goldschmidt, Wilhelm Salomon, Adolph Sauer, Viktor Meyer, P. Lenard, G. Quincke, L. Koenigsberger, and others. He was awarded the degree of Doctor of Philosophy (*summa cum laude*) in December 1900, at the age of twenty-three. While at Heidelberg, Dr. Wright spent one Easter vacation in the shop of Peter Stöe, an outstanding instrument maker who did work for Professor Goldschmidt, including production of two-circle goniometers. This experience with Stöe proved very helpful later in developing the various optical instruments and accessories with which the name of F. E. Wright has long been associated.

After his return to the United States, Dr. Wright was instructor in petrology at the Michigan College of Mines (now the Michigan Institute of Mining and Technology) at Houghton, from 1901 to 1904. He then became associated with the U. S. Geological Survey for two years. In 1906 he was appointed as Petrologist at the newly established Geophysical Laboratory of the Carnegie Institution of Washington where he served with distinction for thirty-eight years until his retirement at the age of sixty-seven years in 1944.





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At the Geophysical Laboratory he developed optical methods for the identification of crystalline phases in artificial silicate melts and his publication of *The Methods of Petrographic-Microscopic Research: Their Relative Accuracy and Range of Application* (Carnegie Institution of Washington Publication No. 158, 204 pp., 1911) did much to stimulate the use of petrographic methods in mineralogy, geology, and chemistry. The paper *The Ternary System CaO—Al<sub>2</sub>O<sub>3</sub>—SiO<sub>2</sub>* by G. A. Rankin, with optical study by F. E. Wright (*Amer. Jour. Sci.*, 4th ser., **39**, 1–79, 1915) is a classic and, besides its wide application to mineralogy and geology, it provided a basis for the development of Portland cement.

As Chairman of the Carnegie Institution's Committee on the Study of the Surface Features of the Moon, Dr. Wright spent many summers in residence at Mt. Wilson Observatory between 1924 and 1939. The results were recorded in the Year Book of the Carnegie Institution of Washington. His measurements on the polarization of light from materials on the moon's surface were the first to indicate the kinds of materials found there.

During the first World War, while serving as a Major in the Ordnance Department of the Army, he contributed extensively to the development of American production of optical glass. In World War II he served as Civilian Adviser to the Joint Optics Committee of the Army and Navy Munitions Board. He helped establish an effective expansion program for the production of critically needed optical glass for urgent military needs. For this fine work the Army awarded him the Gold Medal for Exceptional Service. His many friends in the Army will long remember Colonel F. E. Wright.

Dr. Wright was a member of many scientific societies both here and abroad. These include the Petrologists' Club of Washington, Philosophical Society of Washington, Geological Society of Washington (President, 1924), American Academy of Arts and Sciences, American Philosophical Society, American Physical Society, Geological Society of America (Vice President, 1924), Mineralogical Society of America (President, 1941), Optical Society of America (President, 1918–20), and National Academy of Sciences (elected in 1923, served on numerous important committees, was Vice President from 1927 through 1931, and for twenty years was Home Secretary). He was a foreign fellow of the Geological Society (London) and a member of the Mineralogical Society (London) and the Physical Society (London). One of the last honors he received was the Roebling Medal of the Mineralogical Society of America in 1952.

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PRESENTATION OF THE ROEBLING MEDAL OF  
THE MINERALOGICAL SOCIETY OF AMERICA  
TO WILLIAM FREDERICK FOSHAG\*

EDWARD H. KRAUS, *University of Michigan, Ann Arbor, Michigan.*

Our medalist today has a special relationship with and responsibility to the Roebling family. It is, therefore, fitting that we recall that Colonel Washington A. Roebling, in whose honor the medal is named, gave \$45,000 to our endowment fund on February 15, 1926. This gift permitted the Society to expand materially *The American Mineralogist*. It is also well to remember that Colonel Roebling was a connoisseur and ardent collector of minerals. In fact, he had assembled one of the world's finest private collections. He took especial pride in having, as far as possible, typical representatives of all minerals which had been described.

Following the Colonel's death on July 28, 1926, his collection of 16,000 carefully selected specimens was presented to the National Museum of the Smithsonian Institution in Washington. An endowment fund of \$150,000 was generously established by the Colonel's son, John A. Roebling, the income to be used for the acquisition of new material and the preservation of the collection in the high position it had attained. The recipient of the Roebling medal today was requested by Mrs. Roebling to supervise the packing and transporting of the collection from the Roebling residence in Trenton, New Jersey, to the National Museum. Since then it has been his and his associates' responsibility to maintain the collection in the front rank to which Colonel Roebling had brought it.

William Frederick Foshag was born at Sag Harbor, New York, March 17, 1894. He attended the University of California, where as an undergraduate he specialized in chemistry. Before receiving his bachelor's degree in 1919, he served for a short period as control chemist for the Riverside Portland Cement Company at the famous limestone and mineral locality at Riverside, California. Graduate studies were also pursued at the University of California. The degree of doctor of philosophy was awarded in 1923.

Professor Walter C. Blasdale was one of Foshag's instructors in chemistry. Professor Blasdale was much interested in minerals and is remembered by mineralogists for his analysis of the new mineral benitoite, which was described by George D. Louderback in 1907. It was, however, the University's distinguished mineralogist, Professor Arthur S. Eakle,

\* Presented at the annual luncheon meeting of the Society which was held on November 10, 1953, at Toronto, Canada.



who aroused in Foshag an intense and abiding interest in minerals. It should be mentioned that Professor Eakle was active in our Society and served as its president in 1925. In geology Dr. Foshag came under the stimulating influence not only of Professor Louderback, but also of Andrew C. Lawson and of John C. Merriam, who later became president of the Carnegie Institution in Washington.

In 1919 Dr. Foshag began his long career at the United States National Museum when he was appointed as assistant curator. In 1929 his rank was advanced to curator, and in 1948 he was promoted to the head curatorship of the department of geology, as a worthy successor to his eminent predecessors George P. Merrill and Ray S. Bassler.

During this period of thirty-four years Dr. Foshag has been an extremely active and efficient investigator. He has made many significant contributions to the advancement of our science for he has published nearly one hundred papers. They reveal his comprehensive knowledge of the various branches of the earth sciences. Thirteen new minerals, which are now recognized as independent species, have been described by him or in association with others. This is no mean accomplishment. Moreover, much time has been spent by him in the field studying famous mineral and mining localities in the United States and Mexico. On these expeditions great quantities of valuable material were collected, which have greatly enriched the mineral and geological resources of the museum. The results of these numerous field studies and of the subsequent laboratory investigations of the collected specimens are given in many important papers. The activities of the Mexican volcano Paricutin were also studied and photographed by him.

For years the excellent collections of cut and uncut gemstones in the National Museum have been greatly admired. To add new and unusual material has been Dr. Foshag's constant endeavor. Today he is recognized as a leading authority on gems. He has long been closely identified with the activities of the Gemological Institute of America. He is a member of its Educational Advisory Board and of the Editorial Board of *Gems and Gemology*, the Institute's quarterly journal. He also participates regularly in the annual programs of the American Gem Society.

Since 1949, with his associates George Switzer, G. W. Josephson, and H. T. Chandler, he has prepared the important annual Review of the Diamond Industry for the *Jeweler's Circular-Keystone*, and the chapter on Gemstones for the *Minerals Yearbook* published by the United States Bureau of Mines. His comprehensive and beautifully illustrated article Exploring the World of Gems, which appeared in the *National Geographic Magazine* in 1950, won world-wide commendation.

In 1946 Dr. Foshag and his associate Edward P. Henderson spent over



four months in Japan supervising the grading, classifying, and evaluating the diamonds, worth \$25,000,000, which had been cached by the Japanese and subsequently uncovered by the occupation forces. Also, while in Japan the production of cultured pearls was carefully studied by him. It must be added that Dr. Foshag has devoted much time to the study and classification of meteorites.

Dr. Foshag is a fellow of the Mineralogical and Geological Societies of America, the Geophysical Union, and the Society of Economic Geology, and a member of the Society for Research on Meteorites, and the Washington Academy of Science. He is also an honorary member of the Geological Society of Mexico. The following tribute from the Geological Institute of Mexico was received a few days ago by our secretary, Professor C. S. Hurlbut, Jr.:

"Dr. Foshag has studied numerous aspects of the mineralogy of Mexico through a third of a century. He has visited the country many times to make collections and to study the geology of outstanding mineral localities. He has acquired an unsurpassed—and probably unequalled—knowledge of the minerals of Mexico. This vast store of information he has transmitted freely and in particular has Dr. Foshag contributed to the training of Mexican mineralogists—frequently going to great pains on their behalf and always giving his valuable time with unstinting cheerfulness."

This tribute is signed by the director of the Institute, Teodoro Flores, and by nine Mexican associates and friends of Dr. Foshag.<sup>1</sup>

Dr. Foshag is a charter fellow of our Society and has served as a councilor from 1925 to 1928, as vice-president in 1931, and as president in 1940.

Mr. President:

I have the high honor to present William Frederick Foshag as the twelfth recipient of the Washington A. Roebling Medal for meritorious achievement in the mineralogical sciences.

<sup>1</sup> Enrique M. Gonzalez, Amadeo Larralde, Luis Flores Covarrubias, Clara Flores Covarrubias, Jenaro Gonzalez R., Eduardo Schmitter, Ariel Hernandez V., J. Martinez Portillo, and A. R. V. Arellano.

## ACCEPTANCE OF THE ROEBLING MEDAL OF THE MINERALOGICAL SOCIETY OF AMERICA

WILLIAM F. FOSHAG, *U. S. National Museum, Washington, D. C.*

This is an award that I deeply appreciate, and I sincerely thank the Mineralogical Society of America for so signal an honor. I can be quite frank in telling you that, had I been consulted, I believe I could have suggested more worthy candidates, a situation which can only intensify my gratitude to you.

Professor Kraus mentions that I enjoyed a somewhat favored relationship with Washington Roebling. Indeed, as a young mineralogist, I had the happy opportunities of friendly intercourse with him, both through correspondence and as an invited guest at his home. I can, perhaps, do no better on this occasion than to recall some little-known facets of this remarkable man, particularly since a new generation of mineralogists has come forward since Roebling's death in 1926.



WILLIAM FREDERICK FOSHAG

Recipient of the Roebling medal of The Mineralogical Society of America.

Washington A. Roebling's reputation as an engineer is well known. Several biographies, and numerous newspaper and magazine articles have described his career in this field, and its culmination in the epic achievement of the Brooklyn Bridge. A portrait bust of him on the bronze doors designed for the U. S. Capitol by the sculptor, Louis Amateis, upon which geology is represented by Dana, chemistry by Gibbs, and physics by Henry, symbolizes his place in the cultural development of America.

In addition to being a great engineer, one could class him as a great captain of industry. For many years he was president of the vast Roebling Wire Works. The most spectacular achievement of his company, other than their stupendous suspension bridges, was the fantastic North Sea Mine Net of World War I, 27 million feet of wire cable, equipped to carry 100,000 mines, intended to seal the North Sea against the German sea marauders.

Something, too, has been written about his military exploits during the Civil War. He enlisted as a private and by gallant action and bravery upon the battlefields of many campaigns he rose to the rank of colonel. Few men had a more venturesome battle career and his exploits gained him a considerable reputation. In reply to a letter I once wrote to him about a visit to the diabase quarry at Goose Creek, he replied that he knew the place well, for in the battle of Ball's Bluff nearby, he almost lost his life. If we can believe some of the Southern chroniclers, the Yanks were completely extirpated, with the only Southern casualty that of a soldier who shot himself in the foot while hunting rabbits for the pot, after the battle was over. Roebling remarked in his letter that the only specimen he brought back from there was a bullet in the pommel of his saddle, but he escaped being one of the casualties in this disastrous affair.

It was about this time that the use of balloons in military reconnaissance was introduced and Roebling became one of the first military aviators. On one such ascent he spotted the advance of Lee's Armies toward Pennsylvania, a maneuver that culminated in the Battle of Gettysburg. His most famous exploit was his defense of Little Round Top, when he detected Hooker's advance upon that unguarded flank of the Union Army's lines, and with a few of his men wheeled a cannon into position, delaying the Confederate charge until this critical position could be adequately occupied. Military tacticians credit this action with winning this decisive victory, the turning point of the War.

Roebling's military career reads like a Hollywood scenario, and like a cinema hero, he reaped his reward—he married his general's sister.

But what interests us most here is Roebling's career as a mineralogist. His interest in this science was first aroused when he took a course in

mineralogy at Rensselaer Polytechnic Institute. Just when he decided to make a collection of minerals I neglected to ascertain. Having embarked on this project he pursued it with all the enthusiasm and thoroughness that he applied to being a soldier or to building his famous bridge. His goal was a collection as complete as possible, to include not only all species and subspecies, but also representative specimens of all the useless names with which some mineralogists have confused and confounded our science. To achieve this aim he followed the mineralogical literature intently, and thereby acquired a knowledge of our science that many of us might have envied. He wrote to the mineralogists of his day for the new species they described, and obtained much type material in this manner. He accused one famous prolific creator of species of manufacturing names to sell to him. Besides dealers and other collectors he dealt with, if we read his records, thieves and a damned thief, a fool and a scoundrel, the latter a clergyman from Brooklyn. After almost seventy years of constant endeavor, his collection exceeded in completeness the famous public collections of Europe and America. Some idea of its extent is indicated by a few figures: 46 varieties of feldspar, 48 named pyroxenes, 59 named amphiboles, and 64 varieties of mica. Although it contains many superb or unique crystallized mineral and gem specimens, it is essentially, you could say, a mineralogist's collection, in which the scientific investigator was almost certain to find the rarest materials for research or comparison. And the collection was freely available to any scientist who found occasion to use it. In reply to a request from one researcher he replied, "I am always glad to send out rare minerals. It is of no use to keep specimens locked up in dark drawers forever." This enlightened attitude was rare then, even among the scientifically inclined museums.

As you know, the Roebling Collection has been deposited in the Smithsonian Institution. The liberal terms of the gift and a generous endowment by his son, John, permits us to continue Roebling's policies, to gather in everything of mineralogical interest and to make the collection available to all scientific investigators. Larsen used the collection extensively for many of the rarities not obtainable in other collections for his "Microscopic Determination of the Nonopaque Minerals." The 75 varieties of clay minerals in the collection form the basis of much of the recent work on this difficult group, and the many rare uranium minerals are being found invaluable in the current interest in this metal. Some of the specimens have, like an acquaintance of Roebling's, "gone round the world, around and around like a merry-go-round."

This contribution of Roebling's must be evaluated high among the contributions to our science. It could be accomplished only by someone

dedicated to the task, with ample funds to sustain it and a long life to devote to it. I believe that he was as proud of his collection as he was of his famous bridge.

The Roebling Medal is artistically conceived, a beautiful thing. It depicts him from his favorite portrait, a strikingly handsome man in which his kind, gracious and unaffected nature is clearly evident. It would be a desirable object for its artistic merit alone. And the honor that attaches to it is beyond price. That it should befall me to possess it is the greatest honor that I can imagine.



## PRESENTATION OF THE MINERALOGICAL SOCIETY OF AMERICA AWARD TO LOUIS H. AHRENS\*

ESPER S. LARSEN, JR., *U. S. Geological Survey, Washington, D. C.*

*Mr. President, Ladies, and Gentlemen:*

It is a pleasure to a teacher to watch young men and women grow in their science and become the leaders of their group and the promise of the coming generation. Though I have never been a teacher to Dr. Ahrens, I have had the pleasure of seeing him grow from a promising young geochemist to a leader in his field. Dr. Ahrens was trained in South Africa and he received his Doctor of Science degree in chemistry. He early concentrated in geochemistry and especially on the application of spectroscopy to the solution of problems in geochemistry.

He has studied the distribution of some of the rare elements in rocks and minerals, such as the association of thallium and rubidium. He has contributed much to the problem of determining the age of rocks, using chiefly the ratios of strontium to rubidium, and this method of age determination was largely developed by him.

His two papers on ionization potential will prove useful to all geochemists, and his two books on spectrochemical analysis and wavelength tables of sensitive lines are much used by all spectroscopists, especially those working with rocks and minerals.

Much of Dr. Ahren's work has been carried on in South Africa and at the Massachusetts Institute of Technology. He is soon moving to Oxford, England, as reader in Mineralogy. We are sorry to see him leave America, but wish him a happy and successful time in England and shall follow his work with pleasure and profit. We still claim some part of his success!

The Mineralogical Society of America Award is made to Dr. Ahrens because of his contributions to the measurement of geological time and for the particular paper "Measurements of geologic time by the stron-

\* Ahrens, Dr. Louis Herman, Dept. of Geology and Geophysics, Mass. Inst. Tech. (from Jan. 1, 1954; Dept. of Geology and Mineralogy, Oxford University). Born in Pietermaritzburg, Natal, Union of South Africa, April 24, 1918. B. Sc. (Geology and Chemistry) Univ. Natal, 1939; D. Sc. (Chemistry) Univ. Pretoria, 1944. Analytical Chemist, Gov't Met. Lab., Johannesburg, 1940-1945; Senior Chemist 1946. Post Doc. Fellowship, S. Afr. Council Sci. Ind. Res., 1947-48. Res. Assoc., M. I. T. 1949-1950; Assis't. Prof. 1950-1953. Fellow Roy. Soc. Chem. Gt. Brit. and Ireland, Geol. Soc. Am., Mineral. Soc. Am., Geol. Soc. S. Afr. mem., S. Afr. Chem. Inst. and Fin. Geol. Soc. Jubilee Gold Medal (1948), Geol. Soc. S. Afr. Spectrochemical analysis, geological age, geochemistry and cosmochemistry.

tium method" (*Geol. Soc. Am. Bull.*, **60**, 217-266, 1949). When he began his work on the strontium method many geologists thought that the wide distribution of strontium in rocks and minerals would make the method impractical but Dr. Ahrens has shown that lepidolite and some other minerals can be used for age determinations by the strontium method. At first he was obliged to make his determinations without correction for primary strontium but later he has been able to secure isotopic data on the strontium and so to correct for primary strontium. He has lately developed the method so that he can use biotite for age determinations. This makes the method applicable to many more rocks.

In the past eight years Dr. Ahrens has contributed a long list of excellent papers, chiefly on the subjects of geochemistry and spectroscopy. He is a steady worker, intensely interested in his problems, and he finishes his work in published papers. We can look with confidence to the future and expect many contributions from him on the determinations of geologic time and the distribution of the elements in rocks and minerals and other problems in geochemistry

Mr. President, I have the honor and the pleasure of presenting Dr. Louis H. Ahrens for the Third Mineralogical Society of America Award.

## ACCEPTANCE OF THE MINERALOGICAL SOCIETY OF AMERICA AWARD

L. H. AHRENS, *Massachusetts Institute of Technology, Cambridge, Mass.*

*Mr. President, Dr. Larsen, Fellows and Members:*

You must excuse me if I proceed first to the main substance of my remarks—while you may still be inclined to listen—and express my response of appreciation last.

If we lend fairly sensitive ears and eyes to what is said and written about radioactive age determinations we are impressed by the fact that their value is assessed very differently by two rather distinct groups; those who accept age determinations at their face value with *not enough* question, and those who maintain a rather extreme front of skepticism



L. H. AHRENS

Recipient of the Mineralogical Society of America Award.

about all such determinations. My remarks are aimed mainly at the non-believers.

We may grant that much of their skepticism is warranted, as witness the seesaw history of the development of age research. Nor can their misgivings be allayed by the latest utterances and determinations which have been made by those of us who practise age research: true enough, they say, some determinations are reasonable but why so much discordance in the rash of determinations which have appeared during the last few years? At this point we may turn to the offensive. This very state of apparent confusion in many of the determinations is merely a reflection of the tremendous and heartening development of a whole host of new methods which have mushroomed during the past few years, each with the usual allotment of teething troubles and beginner's errors.

It must be remembered too that the development of an age method is usually exasperatingly slow because of the extraordinary variety of disciplines (geology, geochemistry, mineralogy, radioactivity, various analytical procedures including those which deal with isotope analysis, and many more if you wish) which become involved. Rapid and successful development is often hindered solely by lack of liaison. But here, too, we have grounds for fullest optimism because of recent and increasing full participation (rather than interested co-operation) of physicists and physical chemists in age research. I am even aware of physicists of repute (bless their hearts) who, secretly perhaps, have begun to consider themselves geologists or mineralogists: their enthusiasm for age research has even sent them to the field where they have actually been seen collecting specimens! We welcome them with all our hearts, if I may say so on behalf of mineralogists and geologists.

I make no claim to be prescient, but the writing on the wall is unmistakable, as I see it. If the current activity is maintained and if the close liaison which is now developing persists, we are entitled to look forward to an era in which age determinations of excellent quality should abound and which should serve as an indispensable aid in the solution of a host of geological problems of correlation, both on a fairly local scale and on a global scale.

It is no easy task to accurately express my appreciation of the Award. I cannot hide the fact that I felt honored and delighted with the news, but in accepting the Award I wish to do so with humility and with the hope and belief that it will act as the incentive to my research as it is intended.

# PROCEEDINGS OF THE THIRTY-FOURTH ANNUAL MEETING OF THE MINERALOGICAL SOCIETY OF AMERICA AT TORONTO, CANADA

C. S. HURLBUT, JR., *Secretary.*

The thirty-fourth annual meeting of the Society was held on November 9-11, 1953, at the Royal York Hotel, Toronto, Canada. Scientific sessions were held in the morning and afternoon of November 9th and 11th, and in the afternoon of November 10th, at which 83 papers were presented orally. In order to accommodate such a large number of papers two concurrent sessions were held on two occasions.

The annual luncheon of the Society on November 10th was attended by 185 fellows, members and guests. Following the luncheon the twelfth presentation of the Roebling Medal was made to William F. Foshag, and the third presentation of the Mineralogical Society of America Award was made to Louis H. Ahrens. This time was also the occasion of the presentation of the November-December issue of *The American Mineralogist* to Dr. Waldemar T. Schaller and Dr. Clarence S. Ross. This enlarged issue was dedicated to Drs. Schaller and Ross and contained 34 scientific papers written by their friends and colleagues on the U. S. Geological Survey.

Before the beginning of the scientific session on the afternoon of November 10th the Society was addressed by the Retiring President, J. D. H. Donnay, on *Edifices of Periodic Matter*.

The 1953 Council of the Society met for 8 hours during November 8th and 9th and discussed 20 items of business. The 1954 Council met for 4 hours on November 10th and 11th.

*Recommended Amendment to the Constitution.* When the Constitution was adopted in 1920 there were 48 fellows of the Society. Thus with 6 officers and 4 councilors at large a high percentage of the fellowship was represented on the Council. Through the years the fellowship has gradually increased until in 1953 there were 326 fellows, but the number on the Council has remained the same. In order to bring about a larger representation, the 1954 Council recommends the following amendment to the Constitution of the Society:

## ARTICLE III, SECTION 1\*

The officers of the Society shall be a president, a vice-president, a treasurer, a secretary and an editor, who shall be elected annually. There shall be an executive council consisting of the above officers, the retiring president and [four] fellows at large, *two* to be elected each year for terms of [four] *three* years each.

\* Deletions are indicated by brackets and additions by italics.

## COMMITTEES OF THE MINERALOGICAL SOCIETY OF AMERICA FOR 1954

### *Financial Advisory Committee*

J. P. Marble '54  
E. P. Henderson, *Chairman* '54-'55  
R. E. Fuller '54-'56

### *Auditing Committee*

J. J. Fahey, *Chairman*  
H. F. McMurdie  
W. L. Hill

### *Nominating Committee for Officers*

V. T. Allen, *Chairman*  
R. C. Emmons  
E. H. Kraus

T. N. McVay

J. R. Goldsmith

### *Nominating Committee for Fellows*

A. F. Buddington '54  
H. Inslay '54  
J. T. Lonsdale, *Chairman* '54-'55  
R. B. Ferguson '54-'55  
T. F. Bates '54-'56  
J. J. Runner '54-'56

### *Roebling Medal Committee*

J. W. Gruner, *Chairman*  
R. E. Grim  
L. G. Berry



*Mineralogical Society of America  
Award Committee*

F. J. Turner, *Chairman*  
D. J. Fisher  
J. E. Hawley  
H. W. Fairbairn  
J. Murdoch

*Nomenclature Committee*

C. Frondel, *Chairman* '54  
W. F. Bradley '54  
A. L. Howland '54-'55  
E. W. Nuffield '54-'55  
G. T. Faust '54-'56  
G. Tunell '54-'56

*Publications Committee*

A. F. Buddington, *Chairman* '54  
L. S. Ramsdell '54  
F. A. Bannister '54-'55  
A. Pabst '54-'55

N. L. Bowen '54-'56  
O. F. Tuttle '54-'56

*Board of Associate Editors*

M. Fleischer '54  
G. Tunell '54  
A. Pabst '54-'55  
E. F. Osborn '54-'55  
Ian Campbell '54-'56  
Brian Mason '54-'56

*Representatives*

National Research Council  
J. W. Peoples  
American Geological Institute  
J. P. Marble '54  
E. Ingerson '54-'55  
American Association for the Advancement of Science  
C. S. Hurlbut, Jr.

It should be pointed out that the Roebling Medal Committee, the Mineralogical Society of America Award Committee, the Nominating Committee for Officers, and the Nomination Committee for Fellows make recommendations, but the Council is responsible for final choice. In order to have a larger participation in the affairs of the Society, fellows and members are requested to send to the Secretary suggestions for candidates for Awards and for officers and fellows.

REPORT OF THE SECRETARY

*To the Council of the Mineralogical Society of America:*

ELECTION OF OFFICERS AND FELLOWS

Five hundred and fourteen ballots were cast in the election of officers: 156 by fellows and 358 by members of the Society. The officers elected to serve in 1954 are:

*President:* Sterling B. Hendricks, Bureau of Plant Industry, U. S. Department of Agriculture, Beltsville, Maryland.

*Vice-President:* Harry H. Hess, Princeton University, Princeton, New Jersey.

*Secretary:* C. S. Hurlbut, Jr., Harvard University, Cambridge, Massachusetts.

*Treasurer:* Earl Ingerson, U. S. Geological Survey, Washington, D. C.

*Editor:* Walter F. Hunt, University of Michigan, Ann Arbor, Michigan.

*Councilor (1954-57):* Felix Chayes, Geophysical Laboratory, Washington, D. C.

According to the provisions of the Constitution, the following have been elected to fellowship:

Carl Wellington Beck, University of New Mexico, Albuquerque, New Mexico.

Conrad Burri, Swiss Federal Institute of Technology, Zürich, Switzerland.

Charles Louis Christ, U. S. Geological Survey, Washington, D. C.

Ludmila M. M. Dolar-Mantuani, Hydro Electrical Power Commission of Ontario, Toronto, Canada.

Alfred Joseph Frueh, Jr., University of Chicago, Chicago, Illinois.

Paolo Gallitelli, Istituto di Mineralogia della Università, Modena, Italy.

John Cedric Griffiths, Pennsylvania State College, State College, Pennsylvania.

Norman Fordyce McKerron Henry, University of Cambridge, Cambridge, England.

Riad A. Higazy, University of Cambridge, Cambridge, England.

S. Benedict Levin, Signal Corps Engineering Laboratories, Fort Monmouth, New Jersey.

Dan McLachlan, Jr., University of Utah, Salt Lake City, Utah.

Heinz Meixner, Bergdirektion Hüttenberg, Knappenberg, Kärnten, Austria.

Mary E. Mrose, U. S. Geological Survey, Washington, D.C.

Hans Ramberg, Prinsengata 6a, Trondheim, Norway.

Arthur James Cochran Wilson, University College, Cardiff, Wales

#### CHANGE IN THE BY-LAWS

On the recent ballot, fellows and members were asked to vote on the following proposed change in Article IV, Section 1 of the By-Laws of the Society:

##### *From*

Nominations for office shall be made by the Council. The list shall be published in the journal of the Society at least three months before the annual meeting. Any ten (10) fellows or members may forward to the secretary other nominations for any or all offices. All such nominations reaching the secretary not later than three months prior to the annual meeting shall be printed, together with the names of the nominators, as special ballots. The regular and special ballots shall then be mailed to the general membership. The results shall be announced at the annual meeting, and the officers thus elected shall enter upon duty at the adjournment of the meeting.

##### *To*

Nominations for office shall be made by the Council. The list shall be published in the journal of the Society at least three months before the annual meeting. Any ten (10) fellows or members may forward to the secretary other nominations for any or all offices. All such nominations reaching the secretary not later than three months prior to the annual meeting shall also be printed on the ballots to be mailed to the general membership. The results shall be announced at the annual meeting, and the officers thus elected shall enter upon duty at the adjournment of the meeting.

This change was approved by a vote of 454 in the affirmative with 31 in the negative. It should be pointed out that nominations made by any 10 fellows or members will be indistinguishable on the ballot from nominations made by the Council. However, only *fellows* can be nominated for office.

#### MEMBERSHIP STATISTICS

November 1, 1953

	1952	1953	Gain	Loss
Correspondents	5	4	0	1
Fellows	315	326	15	4
Members	814	857	87	44
Subscribers	799	856	108	51
	<hr/> 1933	<hr/> 2043	<hr/> 210	<hr/> 100

The above figures show a net gain of 11 fellows, 43 members, and 57 subscribers, with a loss of 1 correspondent. Considering the four groups together, there is a gain of 110, giving a total of 2043.

The Society lost through death one of its five correspondents, Paul Niggli of Zürich, Switzerland, the 1947 recipient of the Roebling Medal, and four fellows: Albert A. Klein of the Norton Company, Worcester, Massachusetts; O. Ivan Lee of Jersey City, New Jersey; Ralph L. Rutherford of the University of Alberta, Alberta, Canada; and Frederick E. Wright of Washington, D. C., the 1952 recipient of the Roebling Medal.

Respectfully submitted,  
C. S. HURLBUT, JR., *Secretary*

# REPORT OF THE TREASURER FOR 1953

*To the Council of the Mineralogical Society of America:*

Your Treasurer submits herewith his report for the fiscal year beginning August 1, 1952, and ending July 31, 1953.

## RECEIPTS

Dues and subscriptions.....	\$ 8,323.52
Sale of back numbers.....	1,886.63
Authors' charges on reprints.....	1,190.12
Interest and dividends from endowment.....	4,612.76
Geological Society of America aid for printing the Journal.....	7,845.07
Advertising.....	1,278.18
Sale of Index, volumes 21-30.....	43.10
Sale of Index, volumes 1-20.....	7.00
Aid by author on expense of a paper.....	305.54
Contribution.....	2.63
Contributions toward Ross-Schaller volume.....	1,133.00
	<hr/>
	\$26,627.55
Cash on hand, August 1, 1952.....	3,366.37
	<hr/>
	\$29,993.92

## DISBURSEMENTS

Printing and distribution of the Journal (6 issues).....	\$16,871.99
Printing and distribution of reprints.....	1,407.31
To the Editor, Secretary, and Treasurer.....	1,437.50
Clerical assistance.....	751.25
Postage and express.....	326.39
Printing and stationery.....	252.55
Office supplies and equipment.....	12.90
1952 program and abstracts.....	611.39
Roebing Medal.....	139.32
New securities purchased.....	402.73
Committee expenses.....	5.41
Expenses of officers to 1952 meetings.....	111.30
Safety deposit box.....	7.80
Refunds.....	49.55
Check returned.....	4.00
Telephone and telegraph.....	.81
M. S. A. Award certificate.....	7.50
Reprinting back issue.....	517.95
	<hr/>
	\$22,917.65
Cash on hand, July 31, 1953.....	7,076.27
	<hr/>
	\$29,993.92

The endowment funds of the Society as of July 31, 1953, consist of the following securities:

## BONDS

6M Atlantic Coast Line, 4½.....	\$5,257.50
5M New York Central, 5.....	4,300.00
5M Southern Railway, 5.....	5,743.75
	<hr/>
	\$15,301.25

## PREFERRED STOCKS

200 shares, Southern California Edison, 4.88.....	\$5,250.00
100 shares, Union Pacific, 4.....	4,570.25
60 shares, Jones and Laughlin, A, 5.....	4,987.50
55 shares, United States Steel, 7.....	6,946.20
50 shares, Virginia Electric & Power, 5.....	5,942.50
24 shares, Public Service Electric & Gas.....	728.40
10 shares, Consolidated Edison, 5.....	1,066.64
	<hr/>
	\$29,491.49

## COMMON STOCKS

300 shares, Potomac Electric Power.....	\$4,057.73
200 shares, Greyhound Corporation.....	2,300.00
150 shares, Spencer Kellog.....	3,775.00
100 shares, Columbus & Southern Ohio Electric.....	2,087.50
80 shares, Kroger.....	1,990.00
60 shares, United Fruit.....	3,067.50
56 shares, Standard Oil of New Jersey.....	1,444.84
53 shares, American Telephone & Telegraph.....	7,243.32
50 shares, Chesapeake & Ohio.....	2,368.75
50 shares, Phelps Dodge.....	1,975.00
40 shares, Plymouth Cordage.....	2,050.00
30 shares, U. S. Playing Card.....	2,411.25
	<hr/>
	\$34,770.89
	<hr/>
	\$79,563.63

Respectfully submitted,  
EARL INGERSON, *Treasurer*

## DANA FUND

Disbursements are made to needy mineralogists in war areas and to families of deceased mineralogists in war areas.

## RECEIPTS

Available balance, August 1, 1952.....	\$148.41
Interest.....	.72
	<hr/>
	\$149.13

# DISBURSEMENTS

Disbursed.....	\$ 50.00
Available balance, July 31, 1953.....	\$ 99.13
	<hr/>
	\$149.13

Respectfully submitted,  
EARL INGERSON, *Treasurer*

## REPORT OF THE AUDITING COMMITTEE

*To the President of the Mineralogical Society of America:*

The Auditing Committee has examined and verified the accounts of the Treasurer of the Mineralogical Society of America for the fiscal year beginning August 1, 1952, and ending July 31, 1953. The securities listed in the Treasurer's report, with all future coupons on the coupon bonds attached, are in the safety deposit box at the West End Branch of the Washington Loan and Trust Company in Washington, D. C.

Respectfully submitted,  
ALVIN VAN VALKENBERG, JR.  
SAMUEL ZERFOSS  
HATTEN S. YODER, JR., *Chairman*

## REPORT OF THE EDITOR FOR 1953

*To the Council of the Mineralogical Society of America:*

In presenting the annual editorial report for 1953 at this time, the same general policy will be followed as that used in recent years. With five issues published and distributed and the November-December number emerging from the press, sufficient data are now available to give a reasonably accurate general survey of the year's activity. When the editor's report appears in print in the March-April issue certain details incomplete or not available at the present time will be incorporated so that the final printed report in April can be compared with those given for previous years.

As some of you may have surmised the volume this year will establish a new high level as far as size is concerned. This is due largely to the November-December issue now in press (Ross-Schaller number which contains 34 articles and will run slightly over 400 pages in length; making a total for the year of approximately 1285 pages. This exceeds the previous high mark of 1937 by nearly 100 pages. That volume as you may recall included the very large Palache number, appearing in May 1937. This accomplishment is a matter of great pride to all of us and especial credit is due to Dr. Earl Ingerson and his coworkers for assembling the papers for this special issue, reading proof and looking after many of the details connected with the printing of this number.

Beginning with 1949, by Council action, one issue (May-June number) was set aside for "Contributions to Canadian Mineralogy." This policy was continued in 1953 under the able editorship of Dr. L. G. Berry of Kingston, Ontario, and sponsored by the Walker Mineralogical Club. In this number the guest editor assembled 16 interesting papers on varied mineralogical subjects and assumed full responsibility for their selection and in seeing these contributions through the press. Also it should be recalled that the January-February issue was designated as a "Michigan number."

Thus by Council action 3 of the 6 issues for 1953 were earmarked for specific purposes. Naturally this has caused unusual delay of all papers, long and short, that did not fall within the framework of the purposes designated. I am aware that this may have caused



some slight irritation in certain quarters as the elapsed time between receipt and printing has increased to about a year, instead of the usual six to eight months.

However, it should be stated that the circumstances that caused the delay this year are not apt to occur again in the near future, at least not to the same extent. It is hoped that next year we will be able to return to a more normal and rational printing schedule. And in connection with the printing of our Journal, I believe the Banta Publishing Co. should be commended for their whole-hearted cooperation and their careful, efficient and prompt service in printing our publication. As an indication of this performance, recall the present November-December number which is more than twice the normal size and will be ready for distribution about 30 days before its regular printed schedule.

In summarizing the current volume, as indicated previously, the volume will run about 1285 pages (without index) and contain 93 leading articles. Twenty-three additional short papers, appearing under Notes and News, give an over-all total of 116 published manuscripts for the year. These contributions were received from 135 authors associated with 42 different Universities, research bureaus and technical laboratories. Twenty-three critical book reviews and 13 new minerals described in detail for the first time—faheyite, lipscombite, manganpyrosmaltite, hydrohausmannite, woodruffite, sahamalite, roentgenite, faustite, moraesite, burbankite, calkinsite, hidalgoite and montroseite—are a few of the other items of special mineralogical interest.

As in previous years our contributions were not confined to authors residing in the States. During the current year 22 manuscripts (out of 116) came from 6 countries: Australia, Brazil, Canada, Egypt, England and South Africa, indicating a continued broad service rendered by the Journal.

TABLE 1. DISTRIBUTION OF SUBJECT MATTER IN VOLUME 38

Subjects	Articles	Pages	Per Cent of Total
Leading articles*			
Descriptive mineralogy and paragenesis...	28		
Chemical mineralogy and geochemistry....	21		
Structural crystallography.....	21		
Petrography.....	8		
Memorials.....	4		
Miscellaneous.....	11		
	93	1087	84.7
Shorter articles.....	23	67	
Notes and news.....	41	21	
Proceedings of Societies.....	5	88	
Abstracts of new mineral names.....	2	1	
Book reviews.....	23	20	15.3
Total entries.....	187	1284	100.0
Illustrations.....	318		
Index, Title page, Table of contents.....		25	
Grand total.....		1309	

\* Leading articles averaged 11.7 printed pages each.

This report would not be complete without an expression of deep appreciation for the liberal financial support received from the Geological Society of America toward defraying a substantial portion of the printing costs of the journal. Also the National Science Foundation made a liberal contribution to assist with the printing of the large Ross-Schaller number. For this assistance the Society is indeed grateful for without such aid services of necessity would have to be curtailed.

Following the favorable action by this Council, the editor has written the secretaries of the Mineralogical Societies of Great Britain, France, Switzerland, Germany and Italy suggesting an exchange of advertisements with the thought of stimulating greater interest and possibly resulting in an increase in membership. Sufficient time has not yet elapsed for replies to this suggestion.

And finally a word as to the prospects for 1954. From present indications no depression is in sight, although some recession from this year's top level may be expected. The January-February manuscripts containing 11 main articles and 6 shorter contributions are in the hands of the printer. In addition at the present writing there are on file 51 manuscripts aggregating an estimated 775 pages of typed material, approximately enough for 4 issues, exclusive of the January-February number.

The accompanying Table 1 summarizes in more detail the distribution of subject matter in Volume 38.

Respectfully submitted,  
W. F. HUNT, *Editor*

# LIST OF FORMER OFFICERS AND MEETING PLACES

By recommendation of the Council, a complete list of past officers is printed in the proceedings of the annual meeting of the Society:

## PRESIDENTS

1920 Edward H. Kraus  
1921 Charles Palache  
1922 Thomas L. Walker  
1923 Edgar T. Wherry  
1924 Henry S. Washington  
1925 Arthur S. Eakle  
1926 Waldemar T. Schaller  
1927 Austin F. Rogers  
1928 Esper S. Larsen  
1929 Arthur L. Parsons  
1930 Herbert E. Merwin  
1931 Alexander H. Phillips  
1932 Alexander N. Winchell  
1933 Herbert P. Whitlock  
1934 John E. Wolff  
1935 Clarence S. Ross  
1936 William S. Bayley  
1937 Norman L. Bowen  
1938 Ellis Thomson  
1939 Max N. Short  
1940 William F. Foshag  
1941 Frederick E. Wright

1942 Arthur F. Buddington  
1943 John F. Schairer  
1944 R. C. Emmons  
1945 Kenneth K. Landes  
1946 Paul F. Kerr  
1947 M. J. Buerger  
1948 M. A. Peacock  
1949 John W. Gruner  
1950 George Tunell  
1951 A. Pabst  
1952 Michael Fleischer  
1953 J. D. H. Donnay

## SECRETARIES

1920-1922 Herbert P. Whitlock  
1923-1933 Frank R. Van Horn  
1933-1934 Albert B. Peck  
1934-1944 Paul F. Kerr  
1944- C. S. Hurlbut, Jr.

## VICE-PRESIDENTS

1920 Thomas L. Walker  
1921 Waldemar T. Schaller

1922 Frederick A. Canfield	1942 Martin J. Buerger
1923 George F. Kunz	1943 John W. Gruner
1924 Washington A. Roebing	1944 Harry Berman
1925 Herbert P. Whitlock	1945 George Tunell
1926 George Vaux, Jr.	1946 S. B. Hendricks
1927 George L. English	1947 Carl Tolman
1928 Lazard Cahn	1948 Adolf Pabst
1929 Edward Wigglesworth	1949 J. D. H. Donnay
1930 John E. Wolff	1950 Ralph E. Grim
1931 William F. Foshag	1951 Michael Fleischer
1932 Joseph L. Gillson	1952 J. D. H. Donnay
1933 Frank B. Guild	1953 Sterling B. Hendricks
1934 William A. Tarr	
1935 Ellis Thomson	
1936 Harold L. Alling	
1937 H. V. Ellsworth	
1938 Kenneth K. Landes	
1939 Burnham S. Colburn	
1940 Ian Campbell	
1941 William J. McCaughey	

## TREASURERS

1920-1923 Albert B. Peck
1924-1929 Alexander H. Phillips
1929-1930 Albert B. Peck
1931-1940 Waldemar T. Schaller
1941- Earl Ingerson

## EDITORS

1920-1921 Edgar T. Wherry	1922- Walter F. Hunt
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## COUNCILORS

1920 Arthur S. Eakle, Frank R. Van Horn, Fred E. Wright, Alexander H. Phillips.
1921 Frank R. Van Horn, Fred E. Wright, Alexander H. Phillips, Austin F. Rogers.
1922 Fred E. Wright, Alexander H. Phillips, Austin F. Rogers, Thomas L. Watson.
1923 Alexander H. Phillips, Austin F. Rogers, Thomas L. Watson, Esper S. Larsen.
1924 Austin F. Rogers, Thomas L. Watson, Esper S. Larsen, Arthur L. Parsons.
1925 Thomas L. Watson, Esper S. Larsen, Arthur L. Parsons, William F. Foshag.
1926 Esper S. Larsen, Arthur L. Parsons, William F. Foshag, William A. Tarr.
1927 Arthur L. Parsons, William F. Foshag, William A. Tarr, Alexander N. Winchell.
1928 William F. Foshag, William A. Tarr, Alexander N. Winchell, Ellis Thomson.
1929 William A. Tarr, Alexander N. Winchell, Ellis Thomson, Clarence S. Ross.
1930 Alexander N. Winchell, Ellis Thomson, Clarence S. Ross, Paul F. Kerr.
1931 Ellis Thomson, Clarence S. Ross, Paul F. Kerr, William S. Bayley.
1932 Clarence S. Ross, Paul F. Kerr, William S. Bayley, William J. McCaughey.
1933 Paul F. Kerr, William S. Bayley, William J. McCaughey, Kenneth K. Landes.
1934 William S. Bayley, William J. McCaughey, Kenneth K. Landes, E. P. Henderson.
1935 William J. McCaughey, Kenneth K. Landes, E. P. Henderson, J. F. Schairer.
1936 Kenneth K. Landes, E. P. Henderson, J. F. Schairer, Arthur F. Buddington.
1937 E. P. Henderson, J. F. Schairer, Arthur F. Buddington, Arthur P. Honess.
1938 J. F. Schairer, Arthur F. Buddington, Arthur P. Honess, R. C. Emmons.
1939 Arthur F. Buddington, Arthur P. Honess, R. C. Emmons, Carl Tolman.
1940 Arthur P. Honess, R. C. Emmons, Carl Tolman, D. Jerome Fisher.
1941 R. C. Emmons, Carl Tolman, D. Jerome Fisher, Martin A. Peacock.
1942 Carl Tolman, D. Jerome Fisher, Martin A. Peacock, Adolf Pabst.
1943 D. Jerome Fisher, Martin A. Peacock, Adolf Pabst, C. S. Hurlbut, Jr.

- 1944 Martin A. Peacock, Adolf Pabst, Michael Fleischer, S. J. Shand.
- 1945 Adolf Pabst, Michael Fleischer, S. J. Shand, R. E. Grim.
- 1946 Michael Fleischer, S. J. Shand, R. E. Grim, Joseph Murdoch.
- 1947 S. J. Shand, R. E. Grim, Joseph Murdoch, H. H. Hess.
- 1948 R. E. Grim, Joseph Murdoch, H. H. Hess, Clifford Frondel.
- 1949 Joseph Murdoch, H. H. Hess, Clifford Frondel, Lewis S. Ramsdell.
- 1950 H. H. Hess, Clifford Frondel, Lewis S. Ramsdell, E. F. Osborn.
- 1951 Clifford Frondel, Lewis S. Ramsdell, E. F. Osborn, George T. Faust.
- 1952 Lewis S. Ramsdell, E. F. Osborn, George T. Faust, Victor T. Allen.
- 1953 E. F. Osborn, George T. Faust, Victor T. Allen, C. Osborne Hutton

#### ANNUAL MEETING PLACES

- |                               |                               |
|-------------------------------|-------------------------------|
| 1920 Chicago, Illinois        | 1937 Washington, D. C.        |
| 1921 Amherst, Massachusetts   | 1938 New York, N. Y.          |
| 1922 Ann Arbor, Michigan      | 1939 Minneapolis, Minnesota   |
| 1923 Washington, D. C.        | 1940 Austin, Texas            |
| 1924 Ithaca, New York         | 1941 Boston, Massachusetts    |
| 1925 New Haven, Connecticut   | 1942 No meeting held          |
| 1926 Madison, Wisconsin       | 1943 No meeting held          |
| 1927 Cleveland, Ohio          | 1944 No meeting held          |
| 1928 New York, N. Y.          | 1945 Pittsburgh, Pennsylvania |
| 1929 Washington, D. C.        | 1946 Chicago, Illinois        |
| 1930 Toronto, Canada          | 1947 Ottawa, Canada           |
| 1931 Tulsa, Oklahoma          | 1948 New York, N. Y.          |
| 1932 Cambridge, Massachusetts | 1949 El Paso, Texas           |
| 1933 Chicago, Illinois        | 1950 Washington, D. C.        |
| 1934 Rochester, New York      | 1951 Detroit, Michigan        |
| 1935 New York, N. Y.          | 1952 Boston, Mass.            |
| 1936 Cincinnati, Ohio         | 1953 Toronto, Canada          |

#### RECIPIENTS OF THE ROEBLING MEDAL

- |                                      |                                       |
|--------------------------------------|---------------------------------------|
| Charles Palache, December 1937       | Paul Niggli, December 1947            |
| Waldemar T. Schaller, December 1938  | William Lawrence Bragg, November 1948 |
| Leonard James Spencer, December 1940 | Herbert E. Merwin, November 1949      |
| Esper S. Larsen, Jr., December 1941  | Norman L. Bowen, November 1950        |
| Edward H. Kraus, February 1945       | Fred E. Wright, November 1952         |
| Clarence S. Ross, December 1946      | William F. Foshag, November 1953      |

#### RECIPIENTS OF THE MINERALOGICAL SOCIETY OF AMERICA AWARD

- |                                     |                                     |
|-------------------------------------|-------------------------------------|
| Orville Frank Tuttle, November 1951 | Frederick H. Stewart, November 1952 |
| L. H. Ahrens, November 1953         |                                     |

ABSTRACTS OF PAPERS PRESENTED AT THE THIRTY-FOURTH  
ANNUAL MEETING OF THE MINERALOGICAL SOCIETY  
OF AMERICA, NOV. 9-11, 1953

URANIUM CONTENT OF THE LAVAS OF LASSEN VOLCANIC  
NATIONAL PARK, WASHINGTON

JOHN A. S. ADAMS AND DONALD F. SAUNDERS  
University of Wisconsin, Madison, Wisconsin

Nineteen samples of volcanic rocks from Lassen Volcanic National Park have been analyzed for U, Na, K and Ca. In addition, the alpha activities were measured with scintillometers. U and K correlate positively, indicating that both tend to be enriched as differentiation proceeded toward more silicious lavas. In the samples studied the U is enriched about four fold while the K is enriched about three fold. The correlations with Ca, Na, and total  $K + Na$  are less well defined than the K correlation. The correlation between K and alpha particle activity under standard conditions is rather well defined.

The correlation between alpha particle activity and U is not well defined when all samples are included. However, the samples that are furthest from the main trend may be considered special cases, e.g. mud flows and rocks that were probably partially weathered and thus leached of part of their U. The main trend in the U content vs. alpha particle activity indicates that the Th/U ratio did not shift significantly during the differentiation of the Lassen lavas.

URANIUM CONTENT, ALPHA PARTICLE ACTIVITY, AND  $K_2O, Na_2O, CaO$   
ANALYSIS OF OBSIDIANS, PITCHSTONES, AND TEKTITES

JOHN A. S. ADAMS, DONALD F. SAUNDERS AND EDWARD J. ZELLER  
University of Wisconsin, Madison, Wisconsin

Seventy-four glassy rocks, including obsidians, pitchstones, tektites, and possible tektites have been analyzed for uranium fluorimetrically. Alpha activities have been measured with scintillometers.  $K_2O$ ,  $Na_2O$ , and  $CaO$  have been determined on the flame photometer. The indices of refraction have also been measured. About half of the samples are from the United States. The thermoluminescence of several samples was also measured, but no thermoluminescence was detected except in one obsidian from Ascension Island.

The uranium contents range from 0.77 ppm in an obsidian from Shizuoka Prefecture, Japan, to 16 ppm in an obsidian from Liparii, Italy. Few of these obsidians have as much as 0.50%  $CaO$ . A plot of alpha particle activity under standard conditions vs. uranium content has little scattering from linearity. This linearity indicates that the Th/U ratio is rather constant in the obsidians measured. Several geochemical interpretations of this linearity are advanced.

LOGNORMAL DISTRIBUTION OF THE ELEMENTS

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The statistical nature of the distribution of Li, K, Rb, Cs, Ga, Sc, V, Co, Cr, Cu, F, Pb, La and Zr in many specimens of two specific rock types, granite (New England



and Canada) and diabase (Ontario, Canada) has been examined. Distribution *appears* to be normal only when dispersion is small, as in the examples of Ga (granite and diabase) and Sc, V and Co (diabase only). Skewness, however, increases regularly with increase in the amount of dispersion and is extreme for Cr (granite and diabase) and Sc, V and Pb (granite only). All elements, irrespective of whether they have a small or large dispersion, show a distribution which is *lognormal*, or at least closely approaches it. An assumption of normal distribution leads to completely erroneous predictions about the prevalence of various concentrations of an element, if its dispersion is large—in fact, the occurrence of some granite with negative concentrations of V, Sc, Pb and Cr is predicted on the basis of such an assumption.

An element may show a totally different degree of dispersion in granite and diabase. For example, dispersion of Sc and V is small in diabase but extreme in granite; on the other hand dispersion of Rb and K is small in granite but moderately large in diabase.

These conclusions are based on quantitative spectrochemical determinations, each calibrated with standard rocks.

#### PHACOLITH OF CHARNOCKITIC ADAMELLITE PORPHYRY AT KEIMOES, CAPE PROVINCE, SOUTH AFRICA

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The charnockitic cycle separates the earlier Kheis orogeny from the numerous pegmatites found throughout the Archean of the northwest Cape, dated at 1,200 million years. The phacolith occupies 225 square miles and similar bodies are found elsewhere in the region. Slightly earlier dikes are basaltic and contain labradorite and bronzite phenocrysts and quartzitic xenoliths. Upon emplacement the charnockitic magma contained phenocrysts of andesine and orthopyroxene, blue quartz xenocrysts, and metasedimentary xenoliths. After emplacement augite, hornblende, biotite, clear quartz, titanomagnetite, and potash-soda feldspar crystallized. The magma apparently originated by deep seated contamination of a basaltic magma. Country rocks are psammitic metasediments with minor pelitic and cafemic intercalations, lenses of orthoamphibolite and metagabbro, and syntectonic lenses of tonalitic to granodioritic gneiss. Contacts with the charnockite are sharp when parallel to the foliation of the country rocks and the charnockite becomes finer grained toward such contacts. Cross-cutting contacts are gradational. Migmatitic rocks are adamellite hybrids with charnockitic textures, permeation gneisses, and mixed rocks produced by intermingling of hybrid magma and mobilized country rocks. The magma was emplaced during compression of rigid folded strata, plastically deformed during the Kheis orogeny. After emplacement compression persisted, resulting in the production of compression joints in the charnockite. Differential movements along joints produced cataclastic deformation in adjacent charnockites. Solutions ascended through the joints and altered neighboring charnockite to quartz-epidote rocks. Constituents removed in this process were concentrated below the roof of the phacolith, where the rocks were changed to syenites of pneumatolytic origin.

#### FURTHER DATA ON THE ISOTOPIC COMPOSITION OF COMMON LEAD

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The isotopic composition of about 50 lead minerals of world-wide distribution has been obtained by mass spectrometric analysis. The range in composition lies within the

extremes reported in the literature for common lead. The results indicate a maximum variation of about 4% in the  $\text{Pb}^{207}/\text{Pb}^{209}$  ratio for minerals from the same metallogenic province.

It appears that in most areas a few isotopic analyses of common lead samples will be sufficient to define the abundances to use in correcting a radiogenic lead for contamination by common lead. This in turn virtually eliminates this source of uncertainty in uranium-lead age measurements.

In order to define the age of the earth more accurately, it is necessary to obtain a still larger number of isotopic analyses of common lead from dated localities.

### CALLAGHANITE, A NEW MINERAL

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Studies of small, azure-blue crystals from the working pits of the Gabbs Refractories Inc., Gabbs, Nevada lead to the description of a new hydrated basic copper, magnesium calcium carbonate,  $\text{Cu}_4\text{Mg}_4\text{Ca}(\text{OH})_{14}(\text{CO}_3)_2 \cdot 2\text{H}_2\text{O}$ . The mineral is named callaghanite after Dr. Eugene Callaghan, Director, New Mexico Bureau of Mines and Mineral Resources, in recognition of his geological work on magnesite deposits. The mineral occurs near peridotite dikes intrusive into magnesite and dolomite.

X-ray studies by Weissenberg and precession methods showed callaghanite to be monoclinic; space group either  $C_s^4 - Aa$  or  $C_{2h}^6 - A2/a$ ; and cell dimensions  $a_0 = 8.24 \text{ \AA}$ ,  $b_0 = 11.80 \text{ \AA}$ ,  $c_0 = 10.06 \text{ \AA}$ ,  $\beta = 107^\circ 18'$ . Elements calculated from x-ray data are:  $a : b : c = 0.6983 : 1 : 0.8525$ ;  $p_0 : q_0 : r_0 = 1.2209 : 0.8140 : 1$ ;  $\mu = 72^\circ 42'$ ;  $p'_0 = 1.2788$ ,  $q'_0 = 0.8525$ ,  $x'_0 = 0.3115$ . The unit cell content is 2.

Crystal forms present are  $\{111\}$ ,  $\{\bar{1}11\}$ ,  $\{221\}$ , and  $\{\bar{2}21\}$ . Elements calculated from goniometric data are:  $a : b : c = 0.7252 : 1 : 0.8504$ ;  $p_0 : q_0 : r_0 = 1.2168 : 0.8092 : 1$ ;  $\mu = 72^\circ 5'$ ;  $p'_0 = 1.2788$ ,  $q'_0 = 0.8504$ ,  $x'_0 = 0.3094$ .

Optical data are:  $\alpha = 1.559$ ,  $\beta = 1.653$ ,  $\gamma = 1.680$ ;  $2V = 55^\circ$ ; optically  $(-)$ ;  $Z \wedge c = 18^\circ$ ; pleochroic in blue with absorption  $Z > Y > X$ ;  $r > v$ , strong.

Cleavages are  $\{111\}$  and  $\{\bar{1}11\}$ , perfect; brittle; hardness is  $3-3\frac{1}{2}$ ; transparent habit, pyramidal. Calculated specific gravity is 2.78; measured 2.71.

### NATURE OF CRYSTALLIZATION OF METAMICT MINERALS

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In recent years there has been an increasing interest in the study of metamict minerals with regard to their origin, identification, and possible use as geologic age indicators.

Many of these non-crystalline minerals have been crystallized by heating to comparatively high temperatures and various workers index the x-ray powder patterns of these heated specimens under the assumption that this crystalline material is reformation of the mineral as originally deposited or crystallized in the rock.

It is the intention of this paper to point out that recrystallization to the original structure of the metamict minerals is the exception rather than the rule. Likewise, data will be presented to show that the temperature, and type, of crystallization of some of these minerals are dependent upon their present state of metamictization and upon the present composition which may or may not be their original composition.

It also will be shown that certain related "stable" minerals will dissociate when subjected to the temperatures at which some metamict minerals crystallize.

## ISOTOPIC ABUNDANCES IN ROCKS AND METEORITES

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Hydrogen, nitrogen, and carbon isotopes were studied in a number of rocks and meteorites to clarify some aspects of the problem of the loss of volatiles from the planetary system and of the origin and formation of the atmosphere.

Hydrogen isotopes were analyzed in the water contained in stony meteorites and igneous rocks: the abundance of deuterium in these samples is explainable with single-stage fractionation processes. No nuclear or multiple fractionation processes seem to be involved, and therefore one can conclude that the abundance of hydrogen isotopes does not change remarkably through the planetary system, except for the sun where the deuterium was consumed by nuclear reactions. Particularly interesting are the carbonaceous chondrites, which contain up to 10 per cent of water and show a minimum of contamination by terrestrial water. The percentage difference in the D/H ratio in these meteorites ranges from  $-5\%$  to  $+20\%$  as referred to the average ocean water. Carbon isotopes were also analyzed in these meteorites, showing a high  $C^{13}$  content with respect to the average on the earth and to the ordinary chondrites. Nitrogen isotope ratios have been measured in rocks. Amounts of nitrogen of 0.1 cc are easily extracted and analyzed with the developed technique.  $N^{15}/N^{14}$  variations of some per cent with respect to the atmospheric nitrogen were found.

MAFIC INTRUSIONS AS A CLUE TO THE METAMORPHIC HISTORY OF THE  
SPRUCE PINE DISTRICT, NORTH CAROLINADONALD A. BROBST,<sup>1</sup> ARIE POLDERVAART<sup>2</sup> AND J. LAURENCE KULP<sup>2</sup><sup>1</sup>U. S. Geological Survey, Washington, D.C., and <sup>2</sup>Columbia University, New York, N.Y.

The geologic history of the Spruce Pine district of North Carolina can be traced from the sediments that formed the pre-Cambrian rocks through two periods of regional metamorphism to the intrusion of basaltic dikes, of Triassic (?) age. The sedimentary rocks were recrystallized and plastically deformed during a late Ordovician or early Silurian metamorphic cycle. Ultramafic bodies were intruded early; alaskite and associated pegmatites were emplaced late. Uranium-lead measurements indicate that pegmatites formed  $350 \pm 20$  million years ago. An interval of crustal tension, with brecciation of the rocks, culminated in a Devonian (?) invasion by the gabbro stock and dike swarm now cropping out near Bakersville. In a second period of regional metamorphism (Appalachian ?), both gabbro and cementing material of the breccia were recrystallized. Minerals of the altered gabbro include garnet, hornblende, intermediate plagioclase, and cloudy pyroxene. The older, already recrystallized rocks were essentially unaffected by this period of metamorphism, although chlorite formed in shear zones. Intense plastic deformation was absent in the second cycle. Tension cracks following this cycle were filled by diabasic dikes during the Triassic (?) period.

The first metamorphic cycle was probably characterized by moderate to high temperatures, high water concentration, and relatively high rates of metamorphic reaction, effecting plastic behavior of the rocks. During the second cycle, temperatures were probably the same or slightly higher; but lower water concentration retarded metamorphic reaction, effecting brittle behavior of the rocks. Because of differences in water concentration evidence of two periods of high-temperature metamorphism is not only preserved, but distinguishable.

## GEOCHEMICAL ASPECTS OF INTERSTITIAL MATERIAL IN IGNEOUS ROCKS

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CHARLES R. MCKINNEY,<sup>1</sup> GEORGE J. NEUERBURG,<sup>2</sup> LEON T. SILVER<sup>1</sup> AND AIJI UCHIYAMA<sup>1</sup>

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The chemical composition and lability of "interstitial" material in four rocks have been studied. The Bonsall tonalite from the Southern California batholith yields upon leaching with dilute acid 0.55% leachable material; a granite from Elberton, Georgia, yields 0.39%; a granite from Essonville, Ontario, yields 0.13%; an aplite from the Boulder batholith (Montana) yields 0.091%. The compositions of the material leached from the four rocks with respect to major constituents are remarkably similar. Silica in all cases is low, ranging from 2% to 6%. Major constituents are CaO (15% to 45%), Fe<sub>2</sub>O<sub>3</sub> (11% to 46%) and Al<sub>2</sub>O<sub>3</sub> (20% to 32%). The levels of minor constituents in interstitial material vary widely and are related to the trace element levels in the original rocks and to the chemical natures of the elements.

The rate of removal of interstitial material is dependent upon the hydrogen ion concentration. In 1M acid the interstitial material of the Bonsall tonalite is taken into solution at a rate of 16 per cent per minute; in 0.1M acid the rate of removal is 4.3 per cent per minute at room temperature. Acid is consumed in the solution process and preliminary experiments indicate that the hydrogen ion consumption amounts to 0.01 moles per gram of interstitial material removed.

Experiments have been run for the purpose of determining the effects of particle size distribution upon the liberation of interstitial material.

## TRIMORPHISM IN ZINC SULFIDE

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Experimental evidence was found for the existence of a third polymorphic modification of zinc sulfide. In addition to  $\beta$  zinc sulfide (zincblende or sphalerite) and  $\alpha$  zinc sulfide (wurtzite) structures, a three-layer rhombohedral structure was found to occur in the temperature range of approximately 600° C. to 1020° C. This structure is similar to the zincblende structure referred to hexagonal axes wherein the  $c$  and  $a$  interatomic distances are slightly altered. In conformity with conventional nomenclature it is designated  $\gamma$ -zinc sulfide.

The experimental evidence for the existence of rhombohedral zinc sulfide is the fine structure of cubic (111) x-ray reflections as observed with high resolution spectrometer studies on pure zinc sulfide heated to various temperatures and quenched.

The role of the rhombohedral phase in the sphalerite-to-rhombohedral-to-wurtzite transformation has been studied.

## IRON AND TITANIUM OXIDE MINERALS OF ADIRONDACK ROCKS

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In igneous rocks of the Adirondack region the amount of TiO<sub>2</sub> in ilmenomagnetite (magnetite with exsolved ilmenite) is greatest in some ore segregations in gabbro and anorthosite (12 to 14 per cent), less in hornblende micropertthite granite (5 to 6.7 per cent), and least in micropertthite alaskite (3.0 to 4.3 per cent). The titanium variation



in the ilmenomagnetite of these plutonic rocks parallels that found by Japanese geologists in magnetite of basaltic, andesitic, and rhyolitic volcanic rocks in Japan; it is a function of temperature of formation and affords a wide-range temperature scale. The composition of the ilmenomagnetite, both that accessory in granite and that in ore concentrations in gabbro and anorthosite, leads to an inference of formation at *magmatic* temperatures.

The magnetite of microcline-rich *granitized* rocks, formed at temperatures lower than magmatic, has about 2.2 to 3.1 per cent  $\text{TiO}_2$ . The  $\text{TiO}_2$  in magnetite of gneisses reconstituted in the upper temperature range of granulite-amphibolite facies is 3.1 to 4.1 per cent; in the lower range of amphibolite facies it is only 1 to 2 per cent. Free ilmenite is present in all samples.

Six different oxide assemblages that were analyzed have the *same* ratio of iron to titanium but differ largely in degree of oxidation. The least oxidized combination (magnetite, ilmenite, and (?) ferrous orthotitanate) occurs in olivine gabbro, and the most oxidized facies (rutilohematite and rutile) in a microcline-rich granite gneiss formed from metasediments by potash-rich fluids. The  $\text{Fe}_2\text{O}_3$  in solid solution in magnetite in granite and gneiss decreases with temperature of formation.

#### CRYSTAL STRUCTURE OF BERTHIERITE, $\text{FeSb}_2\text{S}_4$

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The unit cell, space group, and approximate structure of berthierite were reported in earlier communications. The rough structure has now been refined. The observed amplitudes  $|F_{hko}|$  were first put on an absolute basis by Wilson's method which also determined the temperature coefficient,  $B = 1.05$ . The refinement was carried out by using successive Fourier syntheses,  $(F_o - F_c)$  syntheses, and by fitting the Patterson of the proposed structure to the details of the observed Patterson. The final structure has the following parameters: Fe [.314, .335,  $\frac{1}{4}$ ]; Sb<sub>1</sub> [.145, .064,  $\frac{1}{4}$ ]; Sb<sub>2</sub> [.036, .385,  $-\frac{1}{4}$ ]; S<sub>1</sub> [.197, .272,  $-\frac{1}{4}$ ]; S<sub>2</sub> [.428, .186,  $\frac{1}{4}$ ]; S<sub>3</sub> [.229, .494,  $\frac{1}{4}$ ]; and S<sub>4</sub> [.452, .405,  $-\frac{1}{4}$ ]. The new parameters are within a few thousandths of the values determined directly by minimum-function methods. Using these final parameters, a refined absolute  $F_{\text{obs}}$  scale and temperature coefficient were computed. The residual error,  $R = (|F_o| - F_c)/|F_o|$ , is .142 for  $hk0$  and .126 for  $h0l$ .

Each antimony atom is bonded to three sulfur atoms at distances of about 2.5 Å and these  $\text{SbS}_3$  groups share sulfur atoms to form chains running parallel to the  $c$  axis. The iron atoms are surrounded by six sulfur atoms in approximately octahedral arrangement. These octahedra share edges to form chains parallel to the  $c$  axis. The Fe—S distance, however, is about 2.5 Å, which is considerably in excess of that found in structures where the bonds are recognized as covalent (pyrite, 2.26 Å, marcasite, 2.24 Å). The distance is about what would be expected for ionic Fe—S bonds. The structure can therefore probably be regarded as  $\text{Fe}^{++}(\text{SbS}_2)_2^-$ .

#### CRYSTAL STRUCTURE OF LIVINGSTONITE, $\text{HgSb}_2\text{S}_7$

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The unit cell and space group of livingstonite were investigated by Gorman. His triclinic cell has the following dimensions:



$$\begin{aligned}
 a &= 7.650\text{\AA} & \alpha &= 99^\circ 12\frac{1}{2}' \\
 b &= 10.817 & \beta &= 102^\circ 01' \\
 c &= 3.990 & \gamma &= 73^\circ 48'
 \end{aligned}$$

This cell contains 1  $\text{HgSb}_4\text{S}_7$ . Because of the shortness of the  $c$  axis, we thought it might be relatively easy to determine the structure as projected parallel to  $c$ .  $F^2(hk0)$  intensity data were obtained from a  $c$  axis Weissenberg photograph processed by the M.I.T. modification of the Dawton method. With these data a Patterson map was prepared. The one Hg atom can be taken as located at the origin of Gorman's cell. A quantitative interpretation of the peak heights of the Patterson map located the Sb atoms. An  $M_s$  minimum-function map confirmed these locations and delimited areas available for sulfur atoms.

The structure was approached by successive Fourier syntheses. Phases for the  $F_{\text{obs}}(hk0)$ 's were approximated by assuming that the Hg and Sb's determined the phases provided that they accounted for at least  $\frac{1}{3}$  of the full amplitude. Each successive Fourier synthesis revealed one or more sulfur atoms, which were then taken into account when new phases for a next Fourier synthesis were computed.

One of the seven S atoms should occupy an inversion center provided that livingstonite is centrosymmetrical and provided that the formula is correct. We found no S atom in such a position, but did find a somewhat weak pair of peaks in a general position.

The  $xy$  coordinates found are as follows:

	x	y
Hg	0	0
2 Sb <sub>I</sub>	.473	.146
2 Sb <sub>II</sub>	.164	.428
2 S <sub>I</sub>	.243	.180
2 S <sub>II</sub>	.929	.364
2 S <sub>III</sub>	.694	.087
2 S <sub>IV</sub>	.598	.444

This structure investigation is being continued.

#### $\text{C}^{14}$ CONCENTRATION IN MODERN CARBON

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The accuracy of the absolute age of a sample dated by the  $\text{C}^{14}$  method depends on the specific activity of modern carbon.

Measurements have been made on the following contemporary materials: (1) twelve different woods collected on the Lamont grounds; (2) twenty-six air samples selected to show possible variation with physiography, climate, altitude, humidity and time of day; (3) twenty-four shell or associated meat samples; and (4) a number of surface sea water samples.

Previous results indicated that the specific activity was constant for modern wood. A comparison of these data with that obtained in other laboratories shows that within the present assumed 5% error the  $\text{C}^{14}$  concentration in wood, air, surface ocean carbonate, shell and clam or oyster meat is the same (i.e. about 15 dpm/gm.).

If the  $C^{14}$  produced in the upper atmosphere is well mixed within a century, variations in the rate of production with latitude will not affect the  $C^{14}$  ages. The results reported above lend substantial support to this underlying assumption of the method.

## CRYSTAL CHEMICAL STUDIES OF BORATE MINERALS

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For the known members of the series  $2CaO \cdot 3B_2O_3 \cdot xH_2O$ ,  $x = 13, 9, 7$ , and  $5$ . The  $2CaO \cdot 3B_2O_3 \cdot 9H_2O$  is known only as a synthetic mineral. X-ray studies yield the following data:

Mineral	Inyoite	Artificial	Meyerhofferite	Colemanite
Composition	$2CaO \cdot 3B_2O_3 \cdot 13H_2O$	$2CaO \cdot 3B_2O_3 \cdot 9H_2O$	$2CaO \cdot 3B_2O_3 \cdot 7H_2O$	$2CaO \cdot 3B_2O_3 \cdot 5H_2O$
System	Monoclinic	Triclinic	Triclinic	Monoclinic
Space group	$P2_1/a$ ( $C_{2h}^6$ )	$P\bar{1}$ ( $C_i^1$ )	$P\bar{1}$ ( $C_i^1$ )	$P2_1/a$ ( $C_{2h}^6$ )
Z	2	1	1	2
$a_0$	10.63 Å	7.04 <sub>6</sub> Å	6.61 Å	8.743 Å
$b_0$	12.06	9.45 <sub>2</sub>	8.35	11.264
$c_0$	8.40 <sub>3</sub>	6.41 <sub>2</sub>	6.49	6.102
$\alpha$	(90°00')	101°21'	91°00'	(90°00')
$\beta$	114°02'	101°19'	101°31'	110°07'
$\gamma$	(90°00')	99°49'	86°55'	(90°00')
Volume	984.1 Å <sup>3</sup>	400.7 Å <sup>3</sup>	350.5 Å <sup>3</sup>	564.2 Å <sup>3</sup>
Vol./O-atom	20.5 Å <sup>3</sup>	20.0 Å <sup>3</sup>	19.5 Å <sup>3</sup>	17.6 Å <sup>3</sup>

Colemanite seems to contain sheets of oxygen ions linked by boron ions, parallel to the perfect cleavage plane (010). The study of this structure and its relationship to the structures of the other members of the series is being continued.

## CRYSTAL STRUCTURE OF MURDOCHITE

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U. S. Geological Survey, Washington, D.C.

The crystal structure of the new Cu-Pb-O mineral murdochite has been established. It is an ordered NaCl type, approximate formula  $Cu_8PbO_8$ , space group  $Fm\bar{3}m$  ( $O_h^5$ ) with atomic parameters

$$(000; 0 \frac{1}{2} \frac{1}{2}; \frac{1}{2} 0 \frac{1}{2}; \frac{1}{2} \frac{1}{2} 0) +$$

$$4Pb: (a) 000$$

$$8 O_I: (c) \frac{1}{4} \frac{1}{4} \frac{1}{4}; \frac{3}{4} \frac{3}{4} \frac{3}{4}$$

$$24 O_{II}: (e) x00; 0x0; 00x; \bar{x}00; 0\bar{x}0; 00\bar{x} \text{ with } x = \frac{1}{4}$$

The 24 Cu atoms of the cell are distributed statistically over the 28 sites given by:

$$4Cu_{II}: (b) \frac{1}{2} \frac{1}{2} \frac{1}{2}$$

$$24Cu_I: (d) 0 \frac{1}{4} \frac{1}{4}; \frac{1}{4} 0 \frac{1}{4}; \frac{1}{4} \frac{1}{4} 0; 0 \frac{1}{4} \frac{3}{4}; \frac{3}{4} 0 \frac{1}{4}; \frac{1}{4} \frac{3}{4} 0$$

CONTACT METAMORPHISM IN THE CHRISTMAS MOUNTAINS,  
BREWSTER COUNTY, TEXAS

STEPHEN E. CLABAUGH

University of Texas, Austin, Texas

Spurrite, gehlenite, wollastonite, and other high-temperature contact metamorphic minerals occur in a well defined skarn zone at the contact between Lower Cretaceous limestone and gabbro in the central part of the Christmas Mountains. Superimposed

on the high-temperature mineralization is hydrothermal contact metamorphism associated with small, irregular apophyses of pyroxene syenite and nepheline syenite. Irregular stringers of aplitic nepheline analcime syenite in fractures in the upper part of the gabbro appear to be local differentiates of the gabbro. Clinzoisite and zeolites are associated with the nepheline syenite, and syenite projections into the skarn are rich in sphene, strongly pleochroic titanite, and schorlomite. Trachyte and rhyolite porphyry dikes cut the gabbro, and larger intrusive masses of porphyry are numerous around the periphery of the gabbro. A few small replacement bodies of fluorite occur in limestone adjacent to some of the larger rhyolite masses.

### ISOTOPIC GEOCHEMISTRY OF HOT SPRINGS

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Many geochemical and geological problems require data on the rate of transfer of volatile materials from the interior to the surface of the earth; it is difficult to discuss the mechanisms involved without some estimate of the amounts of juvenile material being added to the surface at the present time. An attack on this problem has been begun by the method of isotopic analyses of carbon, oxygen, hydrogen, and nitrogen in the gases, waters, and deposits of thermal areas, and in igneous rocks.

Five areas have been studied: Yellowstone Park; Steamboat Springs, Nev.; Lassen Park, Calif.; The Geysers, Calif.; and Larderello, Italy. Yellowstone Park appears to contain the smallest proportion of magmatic components; the isotopic composition of the various phases is entirely in accord with a limestone origin for the carbon and a meteoric supply for the waters. In other areas the isotopic analyses indicate varying proportions of magmatic constituents which appear to be greatest at The Geysers. It appears that such areas can be used as successive approximations to determine whether juvenile carbon has the composition of carbon in igneous rocks or of mean surface carbon; this determination is important because an estimate of the degree of constancy of the rate of supply through geologic time can be derived from it. The data indicate that diamonds do not have the isotopic composition of primary terrestrial carbon.

### QUANTITATIVE SPECTROGRAPHIC ANALYSIS OF ROCKS USING A METHOD OF MUTUAL STANDARDIZATION

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The mutual standard method is based on the assumption that the intensity ratio of two spectral lines is a function only of the concentration ratio of the two emitting elements. Since the major elements compose essentially 100 per cent of common rocks, the formula

$$\%Al_2O_3 = \frac{100}{1 + \frac{\%SiO_2}{\%Al_2O_3} + \frac{\%TiO_2}{\%Al_2O_3} + \frac{\%Fe_2O_3}{\%Al_2O_3} + \dots + \frac{\%K_2O}{\%Al_2O_3}}$$

is used for the determination of alumina and thence the remaining oxides. Oxide ratios

are found by plots (working curves) of  $\frac{\text{Intensity, analysis line}}{\text{Intensity, Al 2652}}$  vs  $\frac{\% \text{ analysis oxide}}{\% Al_2O_3}$ .

The lines used are Al 2652, Mg 2779, Mn 2801, Fe 2929, Si 2987, Ti 3088, Ca 3179, Na 3302, and K4044 Å.

Analysis by this method may be performed on untreated powders in the d.c. arc, and all elements are determined simultaneously. Working curves prepared from chemically analyzed rocks allow determinations to be made over a wide concentration range, and use of standards of a similar nature to the analysis material ensures good accuracy. The relative deviation ( $\% \sigma$ ) of the method is 10 per cent.

A number of minor elements, notably Cu, Co, Mo, Ni, Pb, and Zr may also be determined with the major elements.

Analytical time is less than two hours per triplicate analysis of an unknown rock.

## TEMPERATURE AND THE DISTRIBUTION OF TRACE ELEMENTS

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The trace element composition of a mineral can be considered as an equilibrium condition controlled by the major composition of the mineral, P-T conditions of formation, and the availability of trace elements.

Preliminary investigations have demonstrated that the hornblende, biotite, garnet, and chlorite Cu, Cr, Ni, and V contents decrease with increasing temperature. The major decreases of these elements—several thousand ppm for Cr and Ni, and several hundred ppm for Cu and V—seem to occur at the rock transformation from epidote-amphibolite to amphibolite facies. The hornblende and biotite Ti and Mn content increases with increasing temperature; however, these minerals are volumetrically small in the granulite and magmatic facies and may release Ti and Mn from the area. Zinc content, in the few samples analyzed for zinc, is uniformly low and may indicate release of Zn before epidote-amphibolite facies. Therefore, these facies changes and mineral reactions may be related to certain ore deposits of the indicated elements.

The anorthosite plagioclases may be the source of titanium in titaniferous iron ores associated with anorthosite. Anorthosite plagioclases are notably high in Ti, Fe, and Mg, most of which occurs as minute, opaque, exsolved, rod-like inclusions in the plagioclase. Granulation and recrystallization of the plagioclases are accompanied by notable decreases in Ti and often Fe and Mg in the four anorthosite areas studied.

Work on the trace element behavior in pyroxenes, olivines, muscovites, epidotes, feldspars and on other trace elements is in progress and will be discussed.

## TYUYAMUNITE, CARNOTITE, AND SENGIERITE

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The following data were obtained by Weissenberg and precession methods; all lengths given in Å to  $\pm 0.3$  per cent.

Tyuyamunite,  $\text{Ca}(\text{UO}_2)_2(\text{VO}_4)_2 \cdot 6\text{-}8\text{H}_2\text{O}$ , from Mesa County, Colorado: orthorhombic,  $a = 10.63$  Å,  $b = 8.36$  Å,  $c = 16.96$  Å; aspect  $Pna^*$ ; pseudo  $Pnan$  ( $D_{2h}^6$ );  $Z = 4$ ;  $G = 3.92$  (obs.). Potassium replaces calcium; the Ca/K atomic ratio is 1.57.

Carnotite,  $\text{K}_2(\text{UO}_2)_2(\text{VO}_4)_2 \cdot 2/3\text{H}_2\text{O}$ , from Cane Spring Pass, near Moab, Utah: monoclinic,  $a = 10.47$  Å,  $b = 8.41$  Å,  $c = 6.91$  Å,  $\beta = 103^\circ 40' \pm 10'$ ;  $P2_1/a$  ( $C_{2h}^5$ );  $Z = 2$ ;  $G = 4.70$  (obs.). The cell dimensions of synthetic anhydrous carnotite reported by Sundberg and Sillén, are similar, except  $c = 6.59$  Å. The water of hydration should therefore be located between the structural layers (001). Carnotite twins by reticular pseudo-merohedry (index 3, obliquity  $ca 1^\circ$ ); twin and composition plane (001).

Sengierite,  $\text{Cu}(\text{UO}_2)_2(\text{VO}_4)_2 \cdot 8\text{-}10\text{H}_2\text{O} (?)$ , from Haut Katanga, Belgian Congo, pre-

viously reported by Vaes and Kerr as orthorhombic: monoclinic,  $a = 10.62$  Å,  $b = 8.10$  Å,  $c = 10.11$  Å;  $\beta = 103^\circ 40' \pm 5'$  (gon.);  $P2_1/a$  ( $C_{2h}^8$ );  $Z = 2$ ;  $G = 4.41$  (obs.).

Sengierite gives sharp x-ray reflections, whereas tyuyamunite and carnotite give streaky reflections indicating disorder in their crystal structures—the normal to the uranyl-vanadate layers describes a small-angle cone around the normal to the cleavage (001). This is ascribed to the weak bonding of  $\text{Ca}^{++}$  and  $\text{K}^+$ , a hypothesis supported by similar observations on the analogous phosphates: sharp spots from meta-torbernite from Cornwall, England; streaky reflections from meta-autunite from Lauter, Saxony.

#### **Pb<sup>210</sup> METHOD OF AGE DETERMINATION**

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The age of uranium-bearing mineral can be determined by measuring the ratio of  $\text{Pb}^{206}$  to  $\text{Pb}^{210}$ . The  $\text{Pb}^{206}$  concentration is determined with the mass spectrometer. The  $\text{Pb}^{210}$  (RaD) concentration is measured by scintillation alpha counting the daughter  $\text{Po}^{210}$  (RaF). Calibration of the alpha counting system is achieved by counting the  $\text{Po}^{210}$  derived from known aliquots of radon. The method has several important advantages. The ratio  $\text{Pb}^{206}/\text{Pb}^{210}$  is insensitive to both radon leakage and uranium leaching.  $\text{Pb}^{210}$  ages on minerals from classical uranium localities are in good agreement with the "best" ages derived from other isotopic data.

#### **SEDIMENTARY ORIGIN AND STRATIGRAPHIC EQUIVALENCE OF THE SO-CALLED CRANBERRY AND HENDERSON GRANITES IN WESTERN NORTH CAROLINA**

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The rocks that have been called Cranberry granite and Henderson granite in the metamorphic complex of western North Carolina have been found to be largely sedimentary in origin. There are four specific field relations that favor this conclusion: (1) At least 75 per cent of the outcrops of these rocks show primary compositional banding, (2) these rocks are interbedded with rocks of known sedimentary origin (e.g., Carolina gneiss), (3) intrusive relations between the Henderson or Cranberry and the Carolina are absent, (4) the regional distribution suggests that the Cranberry and Henderson rock is the lower unit in the southwest-plunging synclinorium. This field evidence is supported by a microscopic study of zircons from these rocks that shows the common phases of the Cranberry and Henderson to be mostly sedimentary. Detailed mapping in the northwest corner of the Morganton quadrangle where the Cranberry and Henderson come together shows that along strike they grade from one into the other. The two formations therefore seem to be stratigraphically equivalent and the minor differences in their composition may be attributed to facies changes in the original sedimentary sequence. The Cranberry and Henderson rock apparently underlies the Roan and Carolina complex in the Spruce Pine District and thus is older than that complex rather than younger as previously believed.

#### **CHEMICAL CONSTITUTION OF PLYGORSKITES AND PILOLITES**

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Sepiolite minerals which might be considered as end members of the palygorskite and pilolite series are characterized by significant variations of their chemical com-



position. This may be expressed by the coefficient  $K$  corresponding to the varying ratio of  $MgO : SiO_2$  in the chemical formulae of sepiolites. In the sepiolites studied by the author and other investigators the coefficient  $K$  covers the range from 1.0 to 0.60, i.e. from  $MgO \cdot SiO_2 \cdot 2H_2O$  to  $6MgO \cdot 10SiO_2 \cdot 10H_2O$ . Consequently the composition of the sepiolite minerals can be expressed by a series of formulae having the following  $MgO:SiO_2$  ratios: 1.0, 0.88, 0.75, 0.66, 0.60, 0.50. An end member of the sepiolite series,  $MgO \cdot 2SiO_2 \cdot 2H_2O$  ( $K = 0.50$ ) was originally (1939) assumed by the author to be a hypothetical mineral and was actually discovered in 1949 in the Urals (Gerasimov, Grushko, Chirvinsky). These formulae of the sepiolite series make it possible to explain the chemical constitution of palygorskites and pilolites. These minerals may be considered as products of an isomorphous isovalent replacement



in the molecules of sepiolite minerals with different  $K$  values. The recalculation of molecular quantities in the analyses of palygorskites and pilolites after the replacement of  $Al_{\cdot\cdot}^{\cdot\cdot}$  through  $Mg_{\cdot\cdot}^{\cdot\cdot}$  enables us to demonstrate that the empirical chemical formulae of these minerals correspond quite exactly to the formulae of the sepiolite series. This point of view has been corroborated by  $x$ -ray investigations and by the thermal analysis data.

# COMPOSITIONAL EVOLUTION OF A MAJOR MARBLE UNIT, GRENVILLE SERIES, NORTHWEST ADIRONDACK MOUNTAINS, NEW YORK

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Marble in the Balmat-Edwards area is at least 1800 feet thick and represents about one-sixth of the marble in and one-ninth of the thickness of the exposed Grenville series. A weighted average composition of marble inferred to be least altered includes 50 minerals, chiefly (volume per cent) dolomite, 42; calcite, 18; quartz, 16; diopside, 8; tremolite, 7; serpentine, 4; talc, 2. As oxides this represents (weight per cent)  $SiO_2$ , 28;  $MgO$ , 17;  $CaO$ , 25;  $CO_2$ , 27; other, 3.

Field studies suggest that pre-metamorphic (sedimentary?) amounts of  $MgO$  and  $CaO$  approximated present values, but that the mineral composition was (weight per cent) quartz, 15; dolomite, 80; other, 5. Major chemical changes during metamorphism were, therefore, doubling of  $SiO_2$  and appreciable decarbonation. Most calcite probably formed during the (incongruent?) solution of dolomite by water-rich, alkali-silicate, magmatic fluids. These fluids deposited quartz and widely redistributed magnesia, concentrating it into zones of tremolite, anthophyllite, talc, and serpentine. Principal pre-metamorphic accessories postulated are calcite, clay minerals, gypsum, and conceivably pyrite,  $H_2S$ , and methane.

Trace elements in least-altered dolomite beds include (modes of 40 analyses, in ppm) Sr, 200; Mn, 700; Ti, 20; Ba, 4; B, 20; Cu, 2. Comparison with other dolomites—some equally, some less metamorphosed, some roughly contemporary, some much younger—suggest that appreciable strontium diffused out of the dolomite during metamorphism about one billion years ago. One analysis for lead, by Patterson, showed a concentration of 2.14 ppm with an isotopic composition exceptionally high in  $Pb^{207}$ . A possible explanation of the values is that both amount and composition of lead were modified during metamorphism by interchange of constituents in the dolomite with those in associated carbonaceous shales.

## SYNTHESES AND STABILITY OF THE PHLOGOPITES

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A one-layer monoclinic (1M) or a three-layer trigonal (3T) hydroxyl-bearing phlogopite has been synthesized. A curve through the points 840° C.—1,000 psi water vapor pressure, 995°—5,000 psi, 1025°—10,000 psi, 1050°—15,000 psi, 1080°—30,000 psi, and 1105°—75,000 psi mark the upper limit of stability of phlogopite. Above this curve the stable phases are forsterite + leucite + orthorhombic  $\text{KAlSiO}_4$  + vapor. These same phases form metastably at temperatures immediately below the curve, particularly at low pressures.

The phlogopite forms quickly at the higher temperatures, and grows very slowly at the lower temperatures. At the lowest temperatures a completely disordered mica (1Md) grows which is believed to be equivalent to the trioctahedral clay micas. It is not possible to distinguish the 1M and 3T polymorphs on the basis of powder  $x$ -ray diffraction patterns; however, the synthetic phlogopite is not 2M. A natural 2M can be transformed to a 1M or a 3T at high temperatures. The phlogopites from lavas are one-layer monoclinic (1M); however, 1M phlogopites are also formed in metamorphosed limestones. No clear relationship between the temperature of formation and the type of stacking in natural phlogopites has been observed. It has not been ascertained whether the polymorphs are related by a univariant curve of equilibrium or are dependent on growth factors.

Eastonite was synthesized, and its decomposition products are forsterite + leucite + orthorhombic  $\text{KAlSiO}_4$  + spinel + vapor. Mahadevite (1M) is close to eastonite in composition and does not lie midway on the phlogopite-muscovite join.

Phlogopite appears on the liquidus in the synthetic "haplogranite." The formation of phlogopite (and biotite) and the absence of muscovite in lavas are accounted for by their stability curves.

Data have also been obtained on the phlogopite-muscovite, phlogopite-eastonite, and phlogopite-annite joins.

CRYSTAL STRUCTURE OF  $\text{KVO}_3$ 

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Anhydrous potassium metavanadate,  $\text{KVO}_3$ , forms colorless crystals on precipitation from a water solution containing KOH and  $\text{V}_2\text{O}_5$  in the mol ratio of 2 to 1, at a pH of 7 to 8. The crystals are orthorhombic, space group  $Pmab$ , with  $a = 5.69 \text{ \AA}$ ,  $b = 10.82$ ,  $c = 5.20$ , the unit cell containing  $4\text{KVO}_3$ . The structure contains infinite chains stretched along the  $a$  axis, with  $\text{VO}_4$  tetrahedra linked by corners in a manner similar to the silicate tetrahedra in diopside. The structure is analogous to that proposed for  $\text{NH}_4\text{VO}_3$  by Lukesh. The chain linkage is contrasted with the fivefold coordinated chain linkage found in  $\text{KVO}_3 \cdot \text{H}_2\text{O}$ , which forms under the same conditions as  $\text{KVO}_3$ .

## ALTERATION PROCESSES IN MONTROSEITE

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Montroseite,  $(\text{V,Fe})\text{O}(\text{OH})$ , has been shown to have a structure analogous to that of diaspora,  $\text{AlO}(\text{OH})$ . Crystals of montroseite give multiple  $x$ -ray patterns, showing one sharp orthorhombic lattice corresponding to the host crystal, and a pair of diffuse

lattices of similar symmetry and dimensions in parallel orientation. The more prominent of the diffuse phases is interpreted as a metastable form of  $\text{VO}_2$ , resulting from oxidation of the host crystal. The concept of an alteration process involving a migration of ions and electrons through an undisturbed oxygen framework is supported by a Fourier analysis of  $x$ -ray intensities of the sharp and diffuse lattices, and by chemical information. The postulated alteration mechanism is illustrated by certain other examples, notably the alteration of lepidocrocite to maghemite, and goethite to hematite.

#### MURDOCHITE, A NEW COPPER LEAD OXIDE

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Chemical analysis and  $x$ -ray structural studies (see abstract by C. L. Christ and J. R. Clark in this issue) establish as a new species, a copper lead oxide from the Mammoth Mine, Tiger, Arizona. The percentage composition, 67.24  $\text{CuO}$ , 30.53  $\text{PbO}_2$ , 0.17  $\text{Fe}_2\text{O}_3$ , 0.05  $\text{SiO}_2$  and 1.11 Insol. yields the formula  $\text{Cu}_6\text{PbO}_8$ . The specific gravity is 6.465 at  $4^\circ \text{C}$ . The mineral forms in tiny lustrous black octahedra associated with wulfenite, diopside, willemite and fluorite.

The new mineral was found by Percy W. Porter, now deceased, of Cavecreek, Arizona and at his request is named in honor of Professor Joseph Murdoch of the University of California at Los Angeles.

#### RADIOACTIVITY OF THE HELIUM-BEARING FORMATIONS OF THE TEXAS PANHANDLE REGION

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The discovery of radon in some of the helium-bearing natural gas in the Texas Panhandle gas field points out the desirability of determining the distribution and occurrence of radon and associated radium, uranium, and helium in formations containing natural gas. The radon content of about 1,200 producing gas wells has been measured, mostly in the Panhandle field. Concentrations from less than 1 to about  $700 \times 10^{-12}$  curies of radon per liter (S.T.P.) have been observed. The highest concentration of radon usually occurs along the fringes of structural highs. Theoretical analysis of radon measurements under conditions of transient gas flow indicates that the measured radon may originate in the immediate vicinity of the bore in most wells. This result is tentatively confirmed by gamma-ray logs in two wells, but so far it has not been possible to obtain adequate samples of the gas-producing beds to determine the radon source. Samples of spent liquor from the acidizing of one well show great excess of radium over uranium. Small grains of uraniferous asphaltite and radioactive petroleum residues are disseminated in drill samples of dolomite from beds well above and within the gas-producing formations. Radon concentration in the gas is not enough to account for the high helium content. The research is being continued, and comprehensive studies of subsurface geology of this and several other gas fields are in progress.

#### ABUNDANCE OF $\text{S}^{32}$ AND $\text{S}^{34}$ IN SOME NATURAL SOURCES OF SULFUR

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An investigation of the abundance of  $\text{S}^{32}$  and  $\text{S}^{34}$  in natural sulfur compounds has been undertaken as part of a study of the origin of Gulf Coast Sulfur deposits.

The abundance expressed as  $S^{32}/S^{34}$  was measured on sulfur dioxide in a Nier type, single collector, mass spectrometer.

A suite of about 25 samples from the Newgulf Salt dome showed that the  $S^{32}/S^{34}$  ratio for the sulfur from the sulfate was about 1.4% lower than that from elemental sulfur. It has also shown that the ratio  $S^{32}/S^{34}$  is not constant in either the sulfate or the native sulfur, variations of as much as 4% having been found in both forms. The  $S^{32}/S^{34}$  from sulfate in the caprock appears slightly lower than in the salt mass suggesting preferential leaching. At the Newgulf Salt dome the  $S^{32}/S^{34}$  in the  $H_2S$  of the bleedwater, in the pyrite of the caprock and in the native sulfur fell within the same range. This suggests that the last step in the formation of the native sulfur is the oxidation of the  $H_2S$ .

A core from near Barreno, Mexico in which elemental sulfur and gypsum were closely associated showed the  $S^{32}/S^{34}$  ratio from the gypsum to be only 6% lower than the elemental sulfur. Laboratory experiments with the sulfur oxidizing bacteria *Thiobacillus Thiooxidans* showed no fractionation effects.

In sea water sulfate samples from the same location the sulfur isotope ratio is the same at the surface as at a depth of 7,600 feet.

#### THE SYSTEM $FeO-SiO_2-H_2O$

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Phase equilibrium relationships are presented for the system  $FeO-SiO_2-H_2O$ . Syntheses are reported for wustite, magnetite, fayalite, greenalite, minnesotaite, grunerite, and hydrated ferro-ferrisilicates of the chlorite and mica families. In a non-oxidizing hydrothermal environment fayalite undergoes serpentinization to form greenalite below 250° C. Greenalite thermally decomposes to fayalite and minnesotaite above 470° C. The upper limit of stability for minnesotaite is placed at 480° C., minnesotaite decomposing to fayalite and quartz with grunerite appearing as a metastable intermediate phase. Fayalite and silica coexist from 480° C. to the limits of experimentation at 925° C. The iron pyroxene, "ferrosilite," was not identified. Distinction is made between the stability relationships observed in a closed, non-oxidizing hydrothermal environment and those to be expected in an open or circulating hydrothermal system.

#### CLASSIFICATION AND CORRELATION OF DIOCTAHEDRAL MICAS AND HYDROUS MICAS

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The method for calculating formulas for montmorillonites from analyses, suggested by Marshall, and the fractional notation used by Ross and Hendricks for writing formulas for montmorillonites is extended to the dioctahedral micas and hydrous micas, which have the same 2-1 layer structure. From formulas so calculated the charges on the tetrahedral and octahedral layers can be calculated. The sum of these charges should agree closely with the sum of the external cations and provide an internal check on the purity of the sample and the accuracy of the analysis.

Such formulas, with a notation of charge on tetrahedral and octahedral layers, provide a means for comparing the composition of micas and hydrous micas and reveal relationships not otherwise evident. From the muscovites and sericites, in which



almost all the charge is on the tetrahedral layer, there is a continuous shift in the relationship between tetrahedral and octahedral charge through the phengites and glauconites to the celadonites, in which the charge is almost wholly octahedral. The micas and hydrous micas can be classified, therefore, according to their tetrahedral-octahedral charge relationship. Such a scheme of classification permits the correlation of specimens containing unusual constituents, such as manganese, vanadium, chromium, etc. And many specimens that have been given specific mineral names are shown to be merely steps in the shifting of the seat of the charge or in the degree of hydration.

# SIMPLE METHOD FOR THE DETERMINATION OF THE PLAGIOCLASE FELDSPARS

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Precise refractive index data on synthetic plagioclase feldspar glasses have been available for over forty years. However, no routine method based upon these data has come into general use for the determination of the natural feldspars. Such a method possesses certain advantages over the many other procedures presently followed. Its theoretical accuracy is about twice that obtainable with established refractive index methods. Index determinations, involving no particular refinements of technique, were made on the glasses of eight chemically analyzed natural plagioclase feldspars. The anorthite contents derived from these data agreed with those calculated from the chemical analyses within about two per cent. The method is simple and rapid, and calls for no costly equipment. It is therefore recommended for inclusion among the standard methods for the determination of the plagioclase feldspars.

# CARIBOU ERUPTIVE COMPLEX, AN INTERPRETATION IN TIME AND SPACE

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The Caribou complex is located in the Canadian shield two miles west of Loring, Ontario, where it intrudes granite-gneisses. Its shape is that of a lopolith, the map picture presenting a funnel tilted to the west and planed off by erosion. Structural, geophysical and petrographic evidence indicates that the magma was emplaced from the east. The complex is mostly composed of norite with local peridotite and pyroxenite facies; granodiorite forms the upper facies of the intrusive. Throughout, the feldspars and pyroxenes of the norite show granulation and mortar structure. They show extensive replacement by hornblende and biotite which are locally intergrown with pyrite, pyrrhotite and titaniferous magnetite. Garnet and quartz form reaction-rims separating feldspar and pyroxene in the norite; in the olivine-bearing rocks hypersthene, spinel and a diopside-spinel symplectite separate the olivine from the feldspar. The intrusive shows cryptic layering with gradual Fe and Na-Si enrichment toward the west on which is superimposed a rhythmic layering of pyroxene and feldspar.

Consolidation of the intrusive was followed by extensive jointing, and granite-pegmatites ascended many of the fractures. Metasomatism of the norite by the pegmatites gave rise to quartz-diorite with a diablastic texture in areas adjoining the pegmatites and may be responsible for the extensive biotite, hornblende and reaction rim formation in the complex. Late-stage faulting broke up the complex into seven blocks. Some of these faults bear nickeliferous pyrrhotite, mineralization thus post-dating the main igneous activity.



## USE OF ZONE OR BAND THEORY IN PROBLEMS OF SULFIDE MINERALOGY

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The metallic or semi-metallic characteristics of many sulfide minerals, demonstrated by such properties as ductility, metallic luster and low electrical resistivity, are evidence of the metallic nature of their chemical bonds. This justifies a limited application of the modern electronic theories of the metallic state to problems involving these minerals.

Brillouin zones of the minerals in the iron-copper-sulfur system have been determined. The volume of these zones and the electron to atom ratios required to fill these zones have been calculated. These minerals exhibit the properties of semi-conductors (increased conductivity with increased temperature) and therefore it is assumed that free electron states exactly fill one of the zones, with a small energy gap between the filled zone and the next higher zone. Use is made of a modification of the electron theory of H. Jones to explain why, as we go from compounds richer in iron and sulfur to compounds richer in copper, we form structures whose corresponding Brillouin zones, when filled, have lower electron to atom ratios.

THERMODYNAMIC RELATIONS AMONG THE VANADIUM OXIDES, AND  
IMPLICATIONS OF THE COLORADO PLATEAU URANIUM-  
VANADIUM DEPOSITS

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Fields of stability of the low-valence vanadium oxides  $V_2O_2$ ,  $V_2O_3$  (VOOH), and  $V_2O_4$  in water solution have been calculated as functions of pH and oxidation potential. The theoretical equilibrium relations indicate that  $V_2O_2$  should not occur naturally; water would decompose, releasing hydrogen, before a sufficiently reducing environment could be attained. The field of  $V_2O_3$  is essentially coexistent with that of  $Fe(OH)_2$ ;  $V_2O_4$  is stable down to approximately the same lower limits of oxidation potential as  $Fe_2O_3$  [ $Fe(OH)_3$ ]. The field of  $V_2O_3$  also is essentially coexistent with the common metal sulfides over a wide range of total sulfur concentration. An implication from the theoretical relations is that the so-called black ores of the Colorado Plateau that contain low-valence uranium and vanadium compounds and metal sulfides (in addition to many other minerals) may be primary and the carnotite-roscelite ores are derived from them by oxidation.

## EQUILIBRIUM THERMAL DECOMPOSITION OF DOLOMITE

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The experimentally determined  $P_{CO_2}$ -temperature stability curve of dolomite rises in an exponential fashion from approximately 400° C. in air to approximately 640° C. at 65 atm.  $CO_2$  pressure. The 400° value is in good agreement with the observed decomposition of dolomite under  $CO_2$ -free hydrothermal conditions.

The calcites formed by the decomposition contain Mg in solid solution. The similar occurrence of Mg in the calcites of numerous unweathered carbonate metamorphic rocks suggests that such solid solutions are stable under proper conditions of temperature,  $CO_2$  pressure, and  $Mg^{++}$  activity.

This equilibrium curve has been further verified using magnesian calcite algal material, from which dolomite may be synthesized by solid state reaction. Inasmuch as dolomite is formed from the algal material within the dolomite stability field and MgO by exsolution outside the field, the curve may be delineated particularly well by this technique.

# PSEUDOMORPHOUS ILLITE AFTER BIOTITE

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Near Marysvale, Utah, hydrothermal alteration of the lowest unit of Bullion Canyon latite has produced unusually well-formed illite pseudomorphs after biotite. A series is described which goes from biotite to illite with intermixed chlorite to a final almost pure illite. In the weathered zones a pseudomorphous series from biotite to mixed illite-chlorite and calcite is developed. Pseudomorphs range from 1-4 millimeters in diameter. Early exsolution of iron and late development of epidote occurs. Optical, stain, thermal, and x-ray data support the interpretation given. The paragenesis of hydrothermal clays in alkali-rich volcanic rocks is believed to be from mafic constituents to a mixture of chlorite and illite to illite. On crystal chemical grounds the extension of the sequence to pinite or sericite seems likely.

# ELECTROMAGNETIC SURVEYING OF DRILL HOLES FOR THE DETECTION OF CONDUCTING SULFIDES

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A diamond drill hole will sample only the diameter of the hole and many ore bodies present can be missed readily. In a manner analogous to surface electromagnetic surveying the region up to a radius of about 400 feet about the hole can be surveyed for conducting sulfides by use of special apparatus. The direction in which these sulfides lie as well as some indication of their size can be inferred from the results. The method of testing vertical drill holes is described showing how it is possible to allow for the departure of the hole from the vertical. Testing of angle holes is outlined although field tests have not been made with this method.

Results of field tests in the Sudbury basin in drill holes passing through barren areas and in holes collared in areas of known sulfides are discussed.

The method is useful in reducing the number of drill holes required to explore an area and in indicating the direction that further drilling should be done.

# KYANITE, ANDALUSITE, AND SILLIMANITE IN THE SCHISTS IN BOEHL'S BUTTE QUADRANGLE, IDAHO

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Kyanite, andalusite, and sillimanite are found together in the cordierite-bearing mica schists of the Prichard formation of the pre-Cambrian Belt series exposed in the Boehl's Butte quadrangle in the southern part of the Idaho "panhandle." Microscopic studies of these schists suggest that the following inversions took place: (1) sillimanite  $\rightarrow$  kyanite, (2) kyanite  $\rightarrow$  andalusite, (3) kyanite  $\rightarrow$  sillimanite, (4) andalusite  $\rightarrow$  sillimanite. These inversions are probably related to the fluctuation of tempera-

ture and stresses during the complex regional and thermal metamorphism to which the schists were subjected. In some thin sections all three crystal forms occur side by side, suggesting that the minerals may have crystallized under physical-chemical conditions in which all three can exist in equilibrium. The association epidote-plagioclase ( $\text{An}_{30}$ ) in the calcium-rich beds of the same area suggests that the temperature during the crystallization was close to  $400^{\circ}\text{C}$ .

#### STUDIES OF RADIOACTIVE COMPOUNDS: VII—PHOSPHURANYLITE AND DEWINDTITE

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Phosphuranylite from Urgeirica, Portugal, with indices  $\alpha$  1.669,  $\beta$  1.710 (Na) shows only a trace of lead spectrographically. Rotation and Weissenberg films gave the data: Orthorhombic, *Bmmb*;  $a = 15.85$ ,  $b = 17.42$ ,  $c = 13.76$  Å.

Dewindtite from Kasolo gave a practically identical powder pattern confirming Frondel's (1950) statement that the two minerals are isostructural. The indexed powder pattern of dewindtite gave the cell constants:  $a = 16.00$ ,  $b = 17.62$ ,  $c = 13.66$  Å. Schoep's analysis and measured density (5.03) are in better agreement with the formula  $\text{Pb}_3(\text{UO}_2)_6(\text{PO}_4)_4(\text{OH})_6 \cdot 9\text{H}_2\text{O}$  (calc. G. 4.95;  $Z = 4$ ) than the hitherto accepted formula  $\text{Pb}_3(\text{UO}_2)_5(\text{PO}_4)_4(\text{OH})_4 \cdot 10\text{H}_2\text{O}$  (calc. G. 4.46;  $Z = 4$ ).

The analyses of phosphuranylite in Frondel (1950) do not lead to rational cell contents. The formula  $\text{Ca}_3(\text{UO}_2)_6(\text{PO}_4)_4(\text{OH})_6 \cdot 9\text{H}_2\text{O}$  which is to be expected if the new formula for dewindtite represents the chemistry of that mineral, requires more calcium than is indicated by the analyses in Frondel (1950). It is concluded that the chemistry of the two minerals is uncertain.

#### RECRYSTALLIZATION OF METAMICT ZIRCON

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The recrystallization of metamict zircons has been studied by observing the heat emission and changes in the density and structure during annealing. It has been found: (1) that the heat energy liberated during recrystallization is proportional to the degree of structure damage, (2) that the effect of heat treatment at constant temperature on the density of metamict zircons can not be explained in terms of a single stage reaction, and (3) that during annealing the unit cell dimensions approach those of undamaged zircon more rapidly than the density approaches that of undamaged zircon.

#### GRANULITES, AMPHIBOLITES, MYLONITES, AND ASSOCIATED ROCKS OF THE CUCAMONGA CANYON AREA, SOUTHEASTERN SAN GABRIEL MOUNTAINS, CALIFORNIA

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The oldest rocks of the Cucamonga Canyon area, California, are granulites that have been folded in an apparently homoclinal sequence, striking N. and dipping almost vertically. The granulites, comprising mineral associations characteristic of the granulite facies, are probably sedimentary and volcanic rocks that have been dynamothermally metamorphosed.

The granulites have been altered to amphibolites and mylonites along NNW. dipping thrust planes. In Aurela Ridge belt, the granulite structures remain almost intact and only minor amounts of polymetamorphic rocks are present. South of Aurela Ridge belt, ENE. trending structures prevail, yet relic granulite structures and minerals are recognizable. In a belt hundreds of feet wide north of Aurela Ridge belt, the mylonites and amphibolites strike ENE. and granulite structures are obliterated, although relic granulite minerals are plentiful. Farther north, ENE. trending mylonites and minor amphibolites occur in Sarac Ridge belt  $\frac{1}{2}$  mile wide; traces of granulite minerals present suggest these rocks may have been derived from the granulites.

The amphibolites and mylonites are syngenetic. While the felsic granulites were being deformed cataclastically to form mylonites, the mafic granulites were deformed through neomineralization to form amphibolites. Mineral associations indicate that these polymetamorphic rocks were probably formed by the metamorphism of the granulites under the amphibolite facies conditions complemented by intense shearing stress.

Gneissose quartz diorite north of Sarac Ridge, probably emplaced synkinematically, may represent mobilized "quartz-dioritized" granulites. Small quartz monzonite intrusives in the quartz diorite pluton, emplaced post-kinematically, may have promoted recrystallization of the mylonites.

#### DEMONSTRATION POLARISCOPE

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The demonstration polariscope is constructed of two crossed Polaroids four inches apart and four inches square. An ordinary 60-watt lamp beneath the lower Polaroid serves as a light source. Above the lower polarizer is a stage on which cleavage fragments or mineral slices can be placed and rotated to demonstrate extinction angles, twinning, interference and other phenomena observed in plane polarized light. By inserting polished spheres of crystals between the Polaroids one can observe interference figures as described by Professor Horace Winchell in 1947.

This polariscope has the additional advantage of permitting demonstration of interference figures produced by thin slices of minerals. A three-inch plastic sphere can be slid into position between the polarizers; and, if a suitably oriented slice is placed on the stage, an interference figure is observed to fill the sphere. A mirror placed over the upper Polaroid at  $45^\circ$  enables one to demonstrate interference figures to a class of twenty or more at one time.

#### PETROLOGY OF ENCHANTED ROCK PLUTON, LLANO AND GILLESPIE COUNTIES, TEXAS

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The Precambrian pluton consists of four concentric zones: outer zone (1), medium to coarse leucogranite and granite; intermediate zone (2), medium to coarse granite and quartz monzonite; intermediate central zone (3), coarse quartz monzonite and alkalic granodiorite; and the core (4), fine to medium leuco-quartz monzonite and leucogranite. Seriate porphyritic texture prevails. Rapakivi texture is most abundant in zone (3) and magmatic origin for it is indicated by lack of negative correlation between normative quartz and orthoclase + albite.

The magma was probably uniform prior to emplacement, and concentric zones were



developed during emplacement. Crystallization-differentiation accompanied by filter-pressing of a crystal mesh in which microcline separated as the first stable crystalline phase are responsible for rock types and their distribution. The finer grain of the core is believed due to partial escape of volatiles and loss of heat, the latter due mainly to depletion of the magma reservoir.

Microcline phenocrysts, despite size, maintain a mean length-width ratio of 2:1 throughout all rock zones. Standard deviation of mean length for zones (1) and (2) is abnormally low and for zone (3) is large. Values for the latter zone are probably due to presence of the filter-pressed fraction and late stage magmatic and deuteric activity.

Forces that produced steep regional folds may have been sufficient to draw the magma into the chamber. Arcuation of schlieren in zone (1) suggests the magma rose with a spiral motion.

#### DISTRIBUTION OF RARE ELEMENTS WITHIN GRANITES AND GRANITIC PEGMATITES

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The distribution and mode of occurrence of rare elements in a given igneous rock are governed by several well-known factors, chiefly: (1) composition of the magma, including concentrations of the rare-element ions; (2) pressure-temperature conditions during crystallization of the magma; and (3) size, charge, polarizability, and other properties of the rare-element ions.

During crystallization of a magma under a given set of conditions, a rare-element ion may enter the structure of a common mineral (Ga in feldspar) or a rarer mineral (Cs in beryl); it may enter, or be trapped by, a much less appropriate mineral structure (Sn in muscovite), followed in some instances by transfer to an exsolved mineral; it may be concentrated in the residual fluid until it forms a mineral of its own (Cs in pollucite); or it may remain dispersed in the residual fluid until ultimately it is bound to the surface of the nearest crystal lattice.

The first three mechanisms seem to account satisfactorily for the distribution of Be, Cs, Rb, Tl, Li, Nb, Ta, the rare earths, Sn, Ti, W, Mo, U, Th, and other rare elements in granitic pegmatites and some granites. The fourth mechanism evidently becomes prominent during crystallization of other granitic rocks, especially those formed from magmas of relatively high viscosity. It results in end-stage attachment of numerous rare-element ions to the crystal surfaces of common silicate minerals. The binding is relatively weak, and these ions are readily mobilized if the host granites are altered or weathered. Despite their low concentrations, enormous quantities of rare elements might thus be leached from a single large body of granitic rock.

#### ZONES OF REGIONAL METAMORPHISM IN THE PRE-CAMBRIAN OF NORTHERN MICHIGAN

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Pre-Cambrian rocks are exposed throughout an area of about 7,500 square miles in northern Michigan. Of this area, approximately 3,300 square miles is underlain by



unmetamorphosed upper pre-Cambrian (Keweenaw), about 2,700 square miles by metamorphosed middle pre-Cambrian (Huronian), and about 1,500 square miles by metamorphosed lower pre-Cambrian ("Archean"). Flat-lying or low-dipping strata of Paleozoic age overlap the pre-Cambrian along the east margin of the area.

The degree of metamorphism is most clearly shown by mineralogic changes in the middle pre-Cambrian sedimentary rocks, especially the iron-formations and the gray-wackes and slates, and in the late middle pre-Cambrian basic intrusive rocks. These rocks were metamorphosed during the Killarney interval of orogeny and minor granite intrusions. At least one earlier period of major metamorphism (Algoman) is recognized and, in nearby areas of Wisconsin and Minnesota, later but areally more restricted metamorphism is related to the intrusion of the upper pre-Cambrian Duluth gabbro and equivalent masses.

Zones of metamorphic intensity delineated by sillimanite, staurolite, garnet, biotite, and chlorite isograds or their equivalents are completely or partly developed around four nodes, two of which lie in immediately adjacent parts of Wisconsin. The zones range in width from less than a mile to about 30 miles, with metamorphism of low grade being areally dominant over that of intermediate or high grade. Retrograde metamorphism, not accompanied by shearing, is widespread.

Granite dikes and small stocks are fairly common in the areas of higher grade metamorphism. Though doubtless both the metamorphism and the granite intrusions were part of the same orogenic cycle, the granite exposed in most areas is clearly later than the regional metamorphism.

#### EXPERIMENTAL STUDIES OF RHEOMORPHISM

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In order to study the successive melting of natural rocks, specimens of granite, syenite, gabbro, diabase, and peridotite were heated to the melting temperature of minerals present. The rock was kept at constant temperature for periods ranging from a few hours to ten days. After quenching, the rock was studied in thin section. Tests were run in air, CO, CO<sub>2</sub>, H<sub>2</sub>O and city gas.

In all tests the ferromagnesian minerals started to melt earlier than feldspar and quartz. In the thin sections the mafic components could be seen converted into glass. At the same temperature the felsic components were unaffected, if we disregard optical changes. Biotite and chlorite seem to be the first minerals to melt.

In a gas furnace the melting starts 200-300° earlier than in an electric furnace and air. In an electric furnace an atmosphere of H<sub>2</sub>O and (or) CO did lower the melting temperature. A reducing atmosphere seems to cause an early melting of minerals containing iron and magnesium. The time of heating did not affect the temperature of melting. The grain size may have a considerable effect.

The sequence of melting in natural rocks thus does not under all conditions correspond to the sequence of crystallization found in artificial melts. The experiments seem to throw some doubts on the theory that aplitic and granitic material is formed by means of partial anatexis. They explain certain structural features in volcanic and plutonic rocks.

# PETROLOGY OF THE ROUND LAKE BATHOLITH AND ITS ASSOCIATED INTRUSIVES

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The Round Lake batholith is a composite, elliptical intrusive extending from Round Lake, south of Kirkland Lake to Matachewan. It is composed of two main phases, a southeastern leucocratic quartz dioritic or trondhjemitic phase and a northwestern granitic phase which is the younger. Both phases are remarkably uniform, massive and structureless except near contacts with older Keewatin rocks. These features together with local chilled contacts and contact breccias are best explained by assuming a magmatic origin for the batholith.

To the north of the batholith lie a series of syenitic stocks of which the Otto Stock, northwest and north of Round Lake has been most completely studied by the author. The core of this stock is very similar in composition and appearance to the granitic phase of the batholith. The major portion of the stock is syenitic, with local marginal phases of coarse pegmatitic texture, and locally, of altered nepheline syenite.

# OPTICAL AND X-RAY STUDIES OF NATURAL HIGH-TEMPERATURE ALKALI FELDSPARS

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Accurate optic axial angle measurements have been made on members of the sanidine-anorthoclase series both in their natural state and after heating at 900° C. for 24 hours. The mean value of the optic angle of feldspars in the compositional range  $Or_{100}-Or_{80}$  decreases and, in the compositional range  $Or_{80}-Or_{37}$ , increases as a result of this heat treatment. Those feldspars which are triclinic at room temperature (anorthoclases) show only a very slight increase in the mean value of the optic angle. X-ray studies show that these changes are related neither to unmixing nor to the variation in optic angle of crystals from the same specimen.

The change in the effect of heat treatment on the optic angle at a composition of  $Or_{80}$  is correlated with the ease with which the change from sanidine to high-sanidine can be accomplished and, at a composition of  $Or_{37}$ , with the change from monoclinic to triclinic symmetry.

# GEOCHEMICAL STUDY OF CHERT AND RELATED DEPOSITS

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Chemical analyses of 24 samples of high-silica sedimentary materials indicate surprisingly similar bulk compositions. Thirteen of the samples are chert; the remainder are novaculite, flint, siliceous oolite, silicified wood, melikaria, and a chalcedony ball from a bauxite deposit. The samples range from Pre-Cambrian to Recent and have a wide geographic distribution.

The narrow range of silica, 95.00-99.95 per cent, shows a facility of sedimentary differentiation to produce pure chemical types. Differences in the minor and trace constituents, however, may reflect different environments and geologic processes.

The following ranges in weight per cent were found for major constituents:  $SiO_2$ , 95.00-99.95;  $Al_2O_3$ , 0.00-1.95;  $Fe_2O_3$ , 0.04-0.90;  $MgO$ , 0.00-0.49;  $CaO$ , 0.00-1.54;  $Na_2O$ ,

0.00-0.09;  $K_2O$ , 0.00-0.33;  $H_2O$ , 0.01-1.03; and  $CO_2$ , 0.00-1.20.  $TiO_2$ ,  $P_2O_5$ ,  $MnO$  and  $Li_2O$  are minor constituents. As and Mo range from less than 1 to 10 p.p.m. Ge is more variable, with an average of about 40 p.p.m. for the 24 samples. The average Ba content is about 350 p.p.m.; Be, Cu, Cr and Sr range from less than 1 to 500 p.p.m. Co, Ga, Ni, Sc, Sn and V are present as traces but Bi, Pb and Zr could not be detected in any of the samples.

#### NEW OCCURRENCE OF LEUCOSPHENITE, IN OIL SHALE FROM UTAH

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Leucosphenite, previously known only from the Narsarssuak, Greenland, pegmatite, is a widespread mineral in the Green River formation of Tertiary age in northeastern Utah. It has been found in several cores from oil wells in Duchesne and Uintah counties, and well cuttings indicate a vertical range of several thousand feet. It occurs in brown oil shale as well-formed crystals several millimeters in length, commonly associated with shortite  $Na_2Ca_2(CO_3)_3$ . It also occurs with analcime-bearing layers in beds of the Green River formation like those described by W. H. Bradley in 1930. The formula of leucosphenite, on the basis of chemical analysis of the Utah mineral and spectrographic analysis of the type Greenland crystals is revised to include 3%  $B_2O_3$  (previously missed) and to omit 3%  $ZrO_2$  (previously reported). The formula tentatively proposed is  $CaBaNa_3BTi_3Si_5O_{29}$ . Morphological and x-ray studies are in progress. The formation of the leucosphenite is believed related to the reaction of volcanic ash with strongly saline solutions as suggested by Bradley for the analcime.

#### SIGNIFICANCE OF ACCESSORY MINERALS IN IGNEOUS ROCKS

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Contrary to the expressed opinion of many petrologists, most of the common accessory minerals (apatite, zircon, sphene, rutile, iron oxides, pyrite) may be expected to crystallize late in igneous rocks. This conclusion is based on the known association of accessory minerals with pegmatites, on the physical-chemical premise that, other things being equal, constituents in small amount should crystallize late in a complex system, and on evidence from ore deposits of apatite, rutile and oxides of iron and titanium.

Studies of the frequency of association of accessory minerals with the various essential constituents of igneous rocks, particularly the basic to intermediate varieties, indicate a marked association with minerals, such as biotite and hornblende, which are low in the reaction series of Bowen. In all types of plutonic igneous rocks, the accessory minerals occur most abundantly along grain boundaries, near fractures and cleavages, or with alteration patches in the essential minerals. These relationships, occasional replacement criteria, and the erratic distribution of accessory minerals in thin sections, bear out the conclusion that most if not all have crystallized in a late (deuteric) stage of the cooling magma.

Conversely, the association of the accessory minerals with biotite and related minerals in acid rocks implies a late origin for this mineral even in these rocks. The more equal distribution of accessory minerals among the essential constituents of paragneisses as compared with igneous rocks offers a criterion for distinguishing paragneisses from those of magmatic origin.

## DISORDER IN THE SUPER-LATTICE OF EKMANITE

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The chemical composition and differential thermal analysis data of ekmanite suggest that this flaky, hydrous-iron-manganese silicate mineral is related to the layer lattice silicates. The results of single crystal  $x$ -ray investigations indicate that ekmanite possesses a structure that is essentially a packing of talc-type sheets and interleaved cations and that the orthohexagonal cell is a multiple of a sub-unit of the following dimensions:  $a = 5.54$  Å,  $b = 9.60$  Å and  $c = 12.08$  Å. The multiplicity of this sub-unit is present along all three axes, resulting in a super-lattice. The distribution of diffuse reflections indicates the presence of a disordered lattice related to random and irregular displacements of the structural layers along the  $a$  and  $b$  axes. This effect can be inferred from the rotation diagrams of ekmanite taken by rotating the crystal around the  $b$  axis which show diffuse streaks along all of the layer lines, and also from normal-beam equatorial Weissenberg photographs taken by rotation around the  $b$  axis which show that the  $h0l$  reflections are spread out along curves of constant  $h$  and continuously varying  $l$  indices.

The chemical composition, DTA curves and the results of the  $x$ -ray investigations suggest a relationship between ekmanite and stilpnomelane.

STRUCTURAL-CHEMICAL CLASSIFICATION OF THE CHLORITES:  
MAGNESIAN CHLORITES

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As a result of the extension of previous hydrothermal studies in the system  $MgO-Al_2O_3-SiO_2-H_2O$  single homogeneous crystalline phases have been synthesized along the chrysotile-amesite join as far as the amesite composition. At lower temperatures phases with the 7 Å antigorite structure are formed.  $X$ -ray data that show the change in unit cell dimensions accompanying the addition of alumina along the join are given. A single phase of the amesite composition is formed only above 300° C., while below this temperature boehmite + an aluminous serpentine form. Above 450° C. all compositions more aluminous than chrysotile yield a more stable 14 Å phase, presumably with the chlorite structure. The equilibrium decomposition temperatures of the 14 Å phases have been determined. Experiments with natural materials indicate that these, too, probably undergo the antigorite-chlorite dimorphism.

These results substantiate Tschermak's concept of an isomorphous series between serpentine and amesite. The dimorphism between the antigorite and chlorite structures suggests a new structural-chemical classification for at least the magnesian chlorites.

## STUDIES OF MINERAL SULPHO-SALTS: XVIII—PAVONITE, A NEW MINERAL

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Alaskaite (Koenig, 1881) from the Alaska Mine, Colorado which was said to be a Bi sulphide of Pb, Ag and Cu was shown by Thompson (1950) to be a mixture of several minerals including aikinite ( $PbCuBiS_3$ ) and matildite ( $AgBiS_2$ ). A second occurrence in Sur-Lipez province, Bolivia (Ahlfeld with an analysis by Herzenberg, 1926) Thompson identified as benjaminite by  $x$ -ray powder patterns.



Single crystal studies of more favorable material show that despite a marked similarity between the powder patterns of type benjaminite from Nevada and the Bolivian mineral, the two are distinct species:

Nevada	a' 13.34	b' 4.06	c' 20.25 A	$\beta$ 104°
Bolivia	a 13.35	b 4.03	c 16.34 A	$\beta$ 94½°

An  $\alpha$ -ray fluorescence analysis and fusion experiments indicate the composition  $\text{AgBi}_3\text{S}_6$  ( $\text{Ag}_2\text{S} \cdot 3\text{Bi}_2\text{S}_3$ ) which is different from that of known sulpho-salts. The Pb and Cu of Herzenberg's analysis are accounted for by the presence of aikinite and chalcopyrite.

Since the type alaskaite has been discredited and its name and supposed composition are not applicable to the Bolivian mineral, it is proposed to name the new species pavonite from the Latin pavo for peacock, in honor of the late Professor M. A. Peacock.

#### ABUNDANCE OF URANIUM AND THE ISOTOPES OF LEAD IN THE EARTH'S CRUST AND METEORITES

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Data have been obtained for the abundances of uranium and the isotopes of lead in basaltic and oceanic materials. The isotopic compositions of lead from the Columbia River basalt and of recent lead from the Pacific Ocean are  $206/204 = 18.12$ ,  $207/204 = 15.45$ ,  $208/204 = 38.08$ , and  $206/204 = 19.04$ ,  $207/204 = 15.69$ ,  $208/204 = 39.00$  respectively. A study of uranium and lead in meteorites has been reported by Patterson, Inghram, Tilton and Brown. The cosmic abundances of uranium and lead are estimated to be  $1 \times 10^{-4}$  atoms and  $8 \times 10^{-3}$  atoms per  $10^4$  atoms of silicon respectively. The composition of primordial lead, if it can be inferred from that in an iron meteorite, is  $206/204 = 9.41$ ,  $207/204 = 10.27$ ,  $208/204 = 28.16$ . It is possible to make the following interpretations from the above data: nearly all of the uranium in the earth is concentrated in the outermost layer of the crust; this concentration was established when the earth was formed; the  $\text{U}^{238}/\text{Pb}^{206}$ ,  $\text{Pb}^{207}/\text{Pb}^{206}$ , and  $\text{Th}^{232}/\text{Pb}^{208}$  ages for the earth's crust have values greater than 4 billion years; basalt may originate from a material from which, during the life of the earth, some uranium has been removed.

#### METAMORPHISM RELATED TO PEGMATITES IN THE GRENVILLE

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Study of wall rocks of pegmatite intrusives near Bancroft, Ontario, indicates the presence of at least two stages of metamorphism:

(1) An older, regional type which produced diopside pyroxenites and amphibolites in lime-magnesium sediments and hornblende-plagioclase gneisses from argillites.

(2) A younger local effect associated with swarms of pegmatite intrusives, appearing as zones of alteration extending to as much as 2 miles from the pegmatite. The alteration appears as metasomatic replacement with the gradual appearance of scapolite, oligoclase, and microcline in the pyroxenites and amphibolites and microcline and quartz in the gneisses.

A general study of pink gneisses in the Grenville indicates that their association with pegmatites is constant; it is concluded that the effects seen near Bancroft are typical.



## STRUCTURE OF MONTMORILLONITE IN RELATION TO THE PHYSICAL PROPERTIES OF BENTONITES

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Properties of bentonites vary with the nature of the montmorillonite mineral and are dependent upon the proportion of exchangeable  $\text{Na}^+$  and  $\text{Ca}^{++}$  ions as well as types of isomorphous substitutions, which, in turn, may be partially related to weathering and near surface alterations in the bentonite deposit.

Theoretical intensity calculations were made for montmorillonite using a multiplicity factor for three possible phase shifts. *X*-ray analyses and theoretical calculations show that naturally occurring, air-dried bentonites contain "intimate mixtures" of three kinds of montmorillonite. One kind has two molecular layers of water, the second only one molecular layer of water, and the third no water at all adsorbed on the layers. "Double water layer particles" contain  $\text{Ca}^{++}$  as the exchangeable base; "single water layer particles" contain  $\text{Na}^+$  as the exchangeable base and "zero water layer particles" are probably electrostatically neutral.

Drilling mud yield values of bentonite samples are correlated with data obtained from the (001) diffraction lines. Best yield values come from bentonite samples with a (001) 'd' value between 12.85 Å and 12.5 Å and with asymmetry values between .4 and .8. Samples with very symmetrical (001) peaks are very pure in one component and show poor yields. A certain amount of exchangeable  $\text{Ca}^{++}$  is needed in a clay-water system to produce a good drilling mud because a few "double water layer particles" create enough disorder in the system to allow dispersion to take place more easily.

## SYNTHESIS, STABILITY AND PROPERTIES OF LAYER SILICATE STRUCTURES. I—SERPENTINE-KAOLINITE FAMILY

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Minerals with the serpentine structure have been prepared from the following compositions:  $\text{Mg}_6\text{Si}_4\text{O}_{10}(\text{OH})_8$ ,  $\text{Mg}_5\text{Al} \cdot \text{AlSi}_3\text{O}_{10}(\text{OH})_8$ ,  $\text{Mg}_6\text{Ge}_4\text{O}_{10}(\text{OH})_8$ ,  $\text{Ni}_6\text{Ge}_4\text{O}_{10}(\text{OH})_8$  and  $\text{Ni}_6\text{Si}_4\text{O}_{10}(\text{OH})_8$ . The appropriate compositions in the following systems do not yield any serpentine (or 1:1 layer) structure:  $\text{ZnO-SiO}_2$ ,  $\text{CoO-SiO}_2$ ,  $\text{MnO-SiO}_2$ ,  $\text{Ga}_2\text{O}_3\text{-SiO}_2$ ,  $\text{MnO-GeO}_2$  and  $\text{Ga}_2\text{O}_3\text{-GeO}_2$ . A comparison of the morphologies of the serpentine minerals formed supports the thesis that in structures of this type a "misfit" between the layers is the basic cause for the formation of cylindrical crystals. Other data show the secondary importance of the presence of foreign ions, temperature of formation and duration of run in conditioning this basic requirement.

A large number of other new phases has also been synthesized, including talc structures from the following systems,  $\text{MgO-GeO}_2$ ,  $\text{NiO-GeO}_2$  and  $\text{NiO-SiO}_2$ , an anthophyllite from the  $\text{MgO-GeO}_2$  system, and a gallia-silica montmorillonite. A value of 8.16% has been determined for the volume expansion accompanying the spinel-olivine structure inversion occurring at 1005° C. in  $\text{Mg}_2\text{GeO}_4$ .

Phase equilibria in the systems  $\text{MgO-GeO}_2\text{-H}_2\text{O}$  and  $\text{NiO-SiO}_2\text{-H}_2\text{O}$  are presented as a set of pressure-temperature curves for the various reactions, and are compared with those for  $\text{MgO-SiO}_2\text{-H}_2\text{O}$ . Comparative data for the various reactions show some unexpected results; thus the stability limits at 10,000 psi water pressure for the serpentines is as follows: (Mg-Si), 490° C.; (Mg-Ge), 520° C.; (Ni-Si), 530° C.; (Ni-Ge), 360° C.

STABILITY RELATIONS OF SOME MINERALS IN THE SYSTEM  $\text{Na}_2\text{O}-\text{Al}_2\text{O}_3-\text{SiO}_2-\text{H}_2\text{O}$ 

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A study has been made of the synthesis of minerals in the low soda portion of the system  $\text{Na}_2\text{O}-\text{Al}_2\text{O}_3-\text{SiO}_2-\text{H}_2\text{O}$  and of the phase equilibria relating these phases with temperature and pressure. Analcime, albite, paragonite, Na-montmorillonite, sodalite, cancrinite, nepheline and a hydrated nepheline phase were the minerals synthesized and studied. The maximum temperature stability of analcime has been located at  $525 \pm 5^\circ \text{C}$ . (from 500 to 20,000 psi) and the minimum temperature of formation of nepheline located at  $460^\circ \text{C}$ . A maximum decomposition temperature for Na-montmorillonite has been found at  $450^\circ \text{C}$ . and for paragonite at  $635^\circ \text{C}$ .

Phase equilibria are shown in a series of pressure-temperature curves separating areas of stability of various phase assemblages.

PRELIMINARY REPORT ON THE SYSTEM  $\text{Na}_2\text{O}-\text{MgO}-\text{SiO}_2$ 

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More than two hundred separate compositions in this system have been prepared and studied by the method of quenching. So far, eight ternary compounds have been recognized, all of which melt incongruently. Several of the new compounds are difficult to identify under the microscope from optical properties, particularly when they are present as tiny crystals which are suitable for rapid attainment of equilibrium between crystals and liquid. All of them are easy to identify by their characteristic x-ray powder patterns. A phase equilibrium diagram is given showing the fields of the several primary phases and the temperatures and compositions of the ternary invariant points.

A knowledge of the crystalline phases and melting relations in this ternary system is prerequisite to studies of more complex hydrous systems which may elucidate the compositions and relationships between alkaline pyroxenes and amphiboles which are major constituents of certain alkaline rocks and minor mineral constituents of many igneous rocks.

## POLYMORPHISM IN ONE DIMENSION

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Lattices of layers of nets of equilateral triangles stacked with the nodes of each net above the centers of the triangles of the net below form an infinite set. Layers are distinguished by their immediate environments as cubic (c) and hexagonal (h). The zinc sulfide structures are examples. An admissible lattice as defined above is described by a linear series as; hcchc. . . . To remove one layer (h) requires energy  $V_1$ ; to remove one layer (c) requires energy  $V_2$ . At equilibrium,  $V, V = V_2 - V_1$ , is related to the absolute temperature and the number of layers of each type;  $n_h/n_c = f(\exp -V/kT)$ . For every value  $n_h/n_c$ , there is a subset of admissible lattices. Each member of the subset is distinguished by a unique value of a parameter  $\sigma$ ;  $\sigma = [n_{pq} - (n_{pp} + n_{qq})] / [n_{pq} + (n_{pp} + n_{qq})]$ ; p and q,  $q = p-1$ , are the two possible distances from h to h in a series and  $n_{pq}$  is the number of contacts of blocks of layers of unlike distance and  $n_{pp}$  and  $n_{qq}$  are the numbers of contacts of like blocks. If  $v_{pq}$ ,  $v_{pp}$ , and  $v_{qq}$ , are the energies associated with each kind of contact,  $v = v_{pq} - \frac{1}{2}(v_{pp} + v_{qq})$ ; v is unit

energy of change of  $\sigma$ ,  $\sigma = f(\exp -v/kT)$ . Admissible lattices fill equal volumes; therefore no volume discontinuities accompany polymorphism in the admissible set and, since no latent heats are observed, these transitions must be of higher order than first.

#### EFFECT OF DIFFUSION ON THE NATURAL ISOTOPIC ABUNDANCE RATIOS

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Some processes of formation and alteration of rocks may have altered the isotopic abundance ratios of the elements in the earth's crust. Although exchange reactions have been used to explain these variations to a great extent, it would appear that isotopic fractionation due to a diffusion process might also explain, at least in part, some of the observed data. The degree of fractionation which can be expected from diffusion is shown from a theoretical study of solid-state and solute-diffusion processes. Solid-state diffusion across a boundary or within a crystal can account for a fractionation of several per cent under certain high-temperature conditions, but under the more usual geological conditions diffusion will cause a fractionation of about 1 per cent or less. Solute diffusion can account for fractionation of about the same order of magnitude. These theoretical results are in qualitative agreement with experimental data in a number of cases.

These studies also show that special sampling precautions should be taken for isotopic-abundance measurements which ultimately will be used to interpret geological processes or age measurements; very small crystals or material leached from crystals might display isotopic abundances which have been altered by diffusion processes.

#### TRACE-ELEMENT BEHAVIOR IN REGIONAL METAMORPHISM

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Sixty-three rocks of the pelitic Devonian Littleton formation of New Hampshire have been analyzed spectrographically for trace elements. The samples represent all grades of metamorphism from shales to sillimanite schists and gneisses. The average concentrations and standard deviations are given as follows in ppm: Ga, 19, 6.3; Cr, 110, 33; V, 120, 39; Li, 110, 104; Ni, 64, 26; Co, 18, 6.6; Cu, 18, 18; Sc, 14, 7.4; Zr, 200, 73; Y, 45, 20; Sr, 710, 310; Pb, 24, 12. In addition, Be, Mo, Sn, La, Ag were sought, but were usually below the sensitivity limits of 30, 10, 20, 100, 1 ppm, respectively. Ba almost always exceeds 1000 ppm.

Figures for individual analyses were grouped according to low, medium, or high grade of metamorphism. Statistical tests show a rather wide range in original composition, which suggests that it is unwise to deduce composition changes from a few analyses. Consideration of the averages, however, shows that the concentration of most elements remained constant during regional metamorphism. Ni and Cu show a slight decrease, and Li and Pb a well-defined increase of the order of 100%, accompanying the metamorphism. This increase is a metasomatism at the trace-element level which probably accompanies the potash-metasomatism encountered by Billings in the same formation.

POTASSIUM ARGON STUDIES AT THE UNIVERSITY OF TORONTO—  
A PROGRESS REPORT

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In 1951-52 the University of Toronto began studies of the radiogenic argon content of potassium minerals in order to obtain a value for the branching ratio of potassium 40, and to investigate the possibility of geological age determinations based on the radioactive decay of potassium. Preliminary results of these studies were sufficiently promising that the project was continued and expanded in the following year. Measurements of the potassium and radiogenic argon content of a number of potassium feldspars differing in age by as much as a factor of five were found to be consistent with a branching ratio of  $0.060 \pm .006$ .

Two new argon measuring apparatuses have been built and the number of research workers has been increased. Present indications are that satisfactory age determinations can be made from argon and potassium measurements in feldspars and micas.

SODIC PLAGIOCLASES: (A) LATTICE PARAMETERS OF THE NATURAL HIGH- AND LOW-TEMPERATURE SERIES AND OF SYNTHETIC CRYSTALS, AND (B) REVISED DETERMINATIVE CHARTS FOR THE NATURAL LOW-TEMPERATURE SERIES

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Natural low-temperature sodic plagioclases are converted into the high-temperature form by prolonged heating near the melting point. Synthetic sodic plagioclases have been crystallized only in the high-temperature form. Accurate measurements of the six (triclinic) lattice parameters have been made on the Philips powder spectrometer of all three types of material from  $An_0$  to  $An_{36}$  using a method similar to that of Donnay and Donnay on the synthetic alkali feldspars. A matrix method giving a least-squares solution for the lattice parameters was used.

The lattice parameters (especially  $\gamma^*$ ) of the low-temperature sodic feldspars vary considerably with composition, thus providing a sensitive method for determining composition by  $x$ -rays (sensitivity  $\sim 0.5\%$  An). For the high-temperature sodic plagioclases only  $\beta^*$  varies appreciably with composition. This parameter is sensitive to a change of 3% An. The difference between the lattice parameters of the two series decreases rapidly as the An content increases to  $An_{35}$ .

A revised determinative chart for the low-temperature sodic plagioclases using the angle between the  $(131)$  and  $(\bar{1}\bar{3}1)$  reflections has been prepared. Poor resolution of the  $(\bar{1}\bar{3}1)$  reflection for compositions in the vicinity of  $An_7$  seriously reduced the precision of the measurement. The angle between  $(241)$  and  $(132)$  is almost as sensitive to differences of composition, however, and has the further advantage of uniformly good resolution over the entire range. A determinative chart has therefore been prepared for these reflections.

THEORETICAL AND X-RAY STUDY OF THE MICA POLYMORPHS

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Th two factors which allow polymorphism in the micas are: (1) the pseudo-hexagonal rings of oxygen atoms around the potassium atoms which link the layers



together, and (2) the staggering of the two silicon sheets which form each layer. There are four possible ways of stacking two adjacent layers. A simple way of depicting the stacking is to give only the positions of the potassium atoms, for the complete structure can be built from this knowledge. Completely random stacking gives a one-layer, monoclinic, disordered structure (1Md). With only one stacking fault in an ordered sequence twin crystals are produced. For ordered stacking an infinity of structures can be built, but the simpler ones are 1M, 2M, 2O (Orthorhombic), 6M, 3T (Trigonal), and 6H (Hexagonal). The theoretical structures have all been found in nature except for the 2O and 6H. Random arrangements of blocks of ordered layers give mixed-layer structures.

Interaxial angles measured on a precession camera agree within the experimental error ( $\pm 5'$ ) with the values for exact geometry for a 1M mahadevite, 2M muscovite (USNM 96460), 3T muscovite from Sultan Basin, Washington, and a 3T phlogopite (USNM R4463). Although bending and absorption make intensity comparisons uncertain, those intensities observed were always consistent with exact symmetry. The same Sultan Basin material had been previously described as 3M on the basis of a finite 2V and an apparent lack of symmetry between equivalent diffuse streaks.

A new twin plane (001) was found in a 1M mahadevite crystal close to eastonite in composition. The (001) twin as well as the previously described [110] twin is demonstrated theoretically. A re-examination of a six-layer, triclinic biotite of Hendricks and Jefferson indicates that this is a 2M mica described on different axes.

## OCCURRENCE OF ALUNITE AND PYROPHYLLITE IN PUERTO RICO

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An alunite-pyrophyllite deposit between the towns of Comerio and Aguas Buenas approximately 20 miles southwest of San Juan, Puerto Rico, has been mapped and studied in detail. The work is being carried on cooperatively by the U. S. Geological Survey and the Puerto Rico Economic Development Administration. The area mapped embraces about 12 square miles, but the principal deposit is limited to an area about 4 miles long and a mile wide along the crest and part way down the flanks of east-west trending Cerro La Tiza Mountain. The alunite is represented principally by finely banded alunite-quartz boulders whose relationship to their source is not definitely understood. It seems probable that some of the boulders, especially the larger ones near the crest are the result of local weathering and have not moved far from their place of origin. The flanks of the mountain below the deposit are composed of deeply weathered greenish-brown volcanic agglomerates most probably of Late Cretaceous age.

Pyrophyllite occurs in foliated outcrops, some as much as 70 feet wide. Small patchy bodies of kaolin group minerals, sericite and alunitic clays occur in exposures along small roads and foot trails through a heavy cover of foliage. Preliminary chemical analyses show that the alunite contains about 6 per cent  $K_2O$  and 2 per cent  $Na_2O$ .

The deposit is believed to be of hydrothermal origin, the solutions having come from quartz diorite batholiths of Eocene age south and southeast of the deposit. The hydrothermal solutions are believed to have come up along a general east-west trending line of weakness. Other smaller deposits of a similar nature have been found at intervals as far as 25 miles eastward along this trend and are being studied.



## PROGRESS IN AIRBORNE RADIOACTIVITY SURVEYING

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Further development of airborne radioactivity surveying by the U. S. Geological Survey and the Oak Ridge National Laboratory during the last 2 years has improved greatly the technique both for prospecting and for geologic reconnaissance. In prospecting for new occurrences of uranium and thorium, local concentrations of radioactive materials of possible economic significance can be detected satisfactorily 500 feet above the ground. For maximum efficiency when using multi-engine aircraft a unit area of at least 50 square miles should be surveyed. Selection of areas for survey should be based on the best geologic inferences drawn from general knowledge of where uranium and thorium are most likely to occur. Promising results have been obtained in areas so selected.

Airborne surveys in North Carolina, Florida, California, and Arizona show that radiation intensity is related to the areal distribution of various rock types; thus such surveys provide a reconnaissance technique for delineating major geologic features with rapidity and at low cost. In geologic reconnaissance, rock types can be distinguished both where rock outcrops occur and where residual soils blanket the bedrock.

Empirical measurements of gamma radiation intensity have been made at various distances over point sources of different gamma radiation energies and over simulated ore outcrops; these data accord with and confirm theoretical analyses of the absorption and scattering of gamma radiation in various media and geometric configurations.

## ISOTOPIC COMPOSITION OF LEAD IN LEAD MINERALS FROM THE COLORADO PLATEAUS

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Approximately 45 specimens of lead minerals have been collected from the Colorado Plateaus and adjacent areas. The isotopic composition of the lead in these minerals has been determined. This work is part of a detailed study of the lead-uranium ages of the uranium ores from the Upper Triassic and Upper Jurassic rocks of the plateaus.

The Pb<sup>208</sup> content of essentially uranium- and thorium-free lead minerals ranges from approximately 24 to 60 per cent. The absence of thorium from the plateaus sediments is reflected by the relatively constant Pb<sup>201</sup>/Pb<sup>208</sup> ratio. Systematic changes in the Pb<sup>208</sup> and Pb<sup>207</sup> contents suggest the addition of small amounts of old radiogenic lead whose age is approximately one billion years.

The systematic variations in the isotopic compositions of the lead minerals from the uranium deposits of the plateaus and the Tertiary deposits adjacent to the Colorado Plateaus strongly suggest a common or similar source for the original lead as well as the added radiogenic lead. The apparent relationship of the lead in the plateaus and adjacent ore deposits in Upper Triassic, Upper Jurassic, and Tertiary rocks is supported by the Tertiary lead-uranium ages which have been determined for these uranium ores.

In addition to the application of the regional variation in the lead isotope abundances to certain problems of ore deposition, the isotopic data on the Plateaus leads may also be used to evaluate the current estimates of the Pb<sup>207</sup>/Pb<sup>206</sup> age of the earth using ore leads. Some of the assumptions required by the Pb<sup>207</sup>/Pb<sup>206</sup> method do not appear to be supported by these new isotopic analyses.

## SYNTHETIC ZINC SULFIDE POLYTYPE CRYSTALS

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X-ray examination of zinc sulfide crystals grown from the vapor phase show them to consist of mixed polymorphic structures in the majority of cases, regardless of their external morphology. In addition to intimate mixtures (on atomic scale) of 2- and 3-layer structures—wurtzite and zinblende (or rhombohedral, Buck and Strock) they also show Frondel and Palache polytypes of 4- and 6- layer structures. Crystals containing 2, 3, 4, 5, 6, 7, 8, 9 and 11 layer structures have been observed. Many crystals show very intense diffuse x-ray reflections from certain *hkl* zones of constant *hk* values, not reported for the natural polymorphs. The role of impurities in determining the structure and growth habit of the crystals has been studied.

## ORDOÑEZITE, ZINC ANTIMONATE, A NEW MINERAL FROM GUANAJUATO, MEXICO\*

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Tin ores from the Santin mine, Guanajuato, Mexico, contain a new mineral having the chemical composition  $\text{ZnSb}_2\text{O}_6$ . The mineral is in small pale to dark brown crystals associated with cassiterite. It is tetragonal,  $a = 4.66$ ,  $c = 9.24$  Å, space group  $P4/mnm$ , specific gravity = 6.64. The structure is the trirutile type, and the mineral is isostructural with tapiolite ( $\text{FeTa}_2\text{O}_6$ ), bystromite ( $\text{MgSb}_2\text{O}_6$ ) and many other artificial antimonates and tantalates. The name ordoñezite is proposed for the mineral after the late Ezequiel Ordoñez, outstanding Mexican geologist.

## USE OF ISOTOPE DILUTION IN DETERMINATION OF GEOLOGIC AGE OF MINERALS

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The method of isotope dilution along with the mass spectrometer for determination of radiogenic  $\text{Sr}^{87}$  and  $\text{Rb}^{87}$  reported by Aldrich, has been greatly extended so that 0.01% of rubidium and 0.0001% of radiogenic  $\text{Sr}^{87}$  have been estimated accurately using only 100 mg. of mineral sample.

For each age determination 0.5  $\mu\text{g}$  of  $\text{Sr}^{90}$  and 0.01  $\mu\text{g}$  of fission rubidium containing 70.2%  $\text{Rb}^{87}$  were used. The possibility of working with such small quantities has greatly simplified chemical separation of strontium and has made separation unnecessary for rubidium. The use of  $\text{Sr}^{90}$ , a readily available fission product absent in natural strontium, makes the interpretation of the mass spectrograms simpler. The radioactivity of  $\text{Sr}^{90}$  and of its daughter isotope  $\text{Y}^{90}$  greatly facilitate chemical manipulation. Determination of ages for minerals containing many times as much normal strontium as radiogenic  $\text{Sr}^{87}$  have been carried out.

Ages are presented for one lepidolite and two biotite minerals. The ages obtained were found to be considerably greater than those found by other methods. Since incorrect ages would result if fractionation of rubidium and strontium had occurred due to weathering, this possibility was studied by determining ages of one biotite after washing with hydrochloric acid and aqua regia. These treatments did not materially affect the value of the ages.

\* Publication authorized by the Secretary, Smithsonian Institution.

DETERMINATION OF POLAR AXIAL ANGLES AND POLAR AXIAL RATIOS FROM INTERFACIAL ANGLES

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A simple method of calculation of crystallographic axial ratios and axial angles, and a simple method of determination of Miller indices from measurements of interfacial angles with the one-circle goniometer or with the contact goniometer, are especially needed by mineralogists and geologists whose principal interests are in branches of geologic science other than crystallography but who wish to make some use of crystallographic data in mineral determination.

In all crystal systems the polar axial angles are defined as the angles between the edges of a parallelepiped constructed with its edges perpendicular to the faces 100, 010, and 001, and with its body diagonal perpendicular to the face 111, and the polar axial ratios are defined as the ratios of the lengths of the edges of this parallelepiped. The edge perpendicular to the face 100 is designated  $p_o'$ , the edge perpendicular to the face 010 is designated  $q_o'$ , and the edge perpendicular to the face 001 is designated  $r_o'$ ; the angle between  $q_o'$  and  $r_o'$  is designated  $\lambda$ , the angle between  $p_o'$  and  $r_o'$  is designated  $\mu$ , and the angle between  $p_o'$  and  $q_o'$  is designated  $\nu$ . The polar axial angle  $\lambda$  is thus by definition the angle between the normals to the faces 010 and 001. Likewise the polar axial angle  $\mu$  is by definition the angle between the normals to the faces 100 and 001, and the polar axial angle  $\nu$  is by definition the angle between the normals to the faces 100 and 010. The polar axial angles of some triclinic minerals can thus be obtained as measured interfacial angles or as the sums of measured interfacial angles.

The polar axial ratios of a triclinic crystal can be calculated by means of the equations:

$$\frac{p_o'}{r_o'} = \frac{l \sin (001 \wedge h0l)}{h \sin (100 \wedge h0l)}, \quad (1)$$

$$\frac{q_o'}{r_o'} = \frac{l \sin (001 \wedge 0kl)}{k \sin (010 \wedge 0kl)}, \quad \text{and} \quad (2)$$

$$\frac{p_o'}{q_o'} = \frac{k \sin (010 \wedge hk0)}{h \sin (100 \wedge hk0)}. \quad (3)$$

The polar axial ratios of some triclinic minerals can thus be obtained as the ratios of the sines of measured interfacial angles.

In the other crystal systems the calculation of the polar elements from interfacial angles is even more simple.

MAGMATIC ORIGIN OF A TERTIARY GRANITE FROM THE ISLAND OF SKYE, SCOTLAND

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Recent detailed study of the properties of quartz and feldspar show that both may yield information as to the conditions under which they crystallized. The inversion temperature of quartz can be used as an indicator of relative temperatures of crystallization; high-temperature and low-temperature feldspars can be distinguished by determinations of composition and optic angle.

Previous investigation showed that quartz samples from a group of granites invert at higher temperatures than do quartz samples from a group of rhyolites, and it was suggested that the observed difference between "rhyolite quartz" and most "granite

quartz" probably results from late-stage recrystallization of granite minerals at low temperature. A small pluton on the Island of Skye was selected for study as a type of occurrence which might yield granite which had not recrystallized completely. Quartz was separated from 25 specimens and the inversion in every case was found to be similar to that of quartz from rhyolites previously studied. The alkali feldspar holds up to 45% albite in solid solution and has properties characteristic of sanidine which has changed partially to orthoclase cryptoperthite. The plagioclase feldspar is high-temperature oligoclase. The granite has the textural features of a granite together with the mineralogical features more commonly found in rhyolite. The evidence thus points to a magmatic origin of the granite.

#### APPLICATIONS AND LIMITATIONS OF GEOPHYSICAL PROSPECTING IN NEW BRUNSWICK

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Employment of a combination of two geophysical methods, the airborne magnetic and the ground electromagnetic, has led to the discovery of two important new ore-bodies in the Bathurst area of New Brunswick. Several other geophysical methods are currently in use in New Brunswick, notably resistivity, ground magnetic and geochemical soil testing. The part which each of the five methods noted above has played in the discovery of sulfide mineralization is noted. The applicability and limitations of each method are described particularly with reference to the problem of distinguishing between sulfide mineralization, graphitic sediments (which are abundant in the area) and magnetite deposits.

The airborne magnetometer maps structure by virtue of its response to magnetite content in rocks and ores. The electrical methods map electrically-conducting bodies such as sulfide mineralization and graphitic sediments. Geochemical soil testing, in the limited number of cases in which it has been applied, has been successful in separating those zones of good electrical conductivity which contain quantities of such metals as copper and zinc from those which do not contain these metals.

#### MINERAL ASSOCIATIONS AND TYPES OF URANIUM ORES ON THE COLORADO PLATEAUS

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Uranium ores from the Colorado Plateaus are classified in two main types: (1) uranium with vanadium, (2) uranium with copper and other metals. Each type is subdivided into highly oxidized or relatively unoxidized ore.

The vanadium-uranium ratio of the vanadiferous ores ranges from about 30:1 at Placerville and Rifle, Colo., to about 1:1 at Temple Mountain, San Rafael district, Utah. The chief uranium minerals of the highly oxidized ore are the uranyl vanadates: carnotite, tyuyamunite, and metatyuyamunite. The other vanadium minerals include roscelite, vanadium hydromica, corvusite, fernandinite, and hewettite, with small amounts of pascoite, hummerite, rossite, metarossite, steigerite, fervanite, hydrated vanadium pentoxide, and the sodium analogue of hewettite, the last two being new minerals whose descriptions will be published soon.

The unoxidized vanadiferous ores are black and contain a new black uranium mineral found in 9 mines, pitchblende, and montroseite and at least 2 other low-valence



vanadium oxides. They are associated with pyrite and small amounts of sulfides (or selenides) of lead, copper, zinc, cobalt, nickel, molybdenum, and silver. Melanovanadite, corvusite, and fernandinite may represent a transition to oxidized ore.

The oxidized nonvanadiferous ore is characterized by bright-colored secondary minerals: about 18 yellow, orange, or green uranium minerals, including hydrated oxides, carbonates, sulfates, phosphates, arsenates, and silicates, as well as many secondary blue or green copper minerals, 5 different pink cobalt "blooms," and traces of molybdenum, lead, zinc, nickel, and silver.

The unoxidized nonvanadiferous ore is also black and contains pitchblende, the new uranium mineral mentioned above, chalcopyrite, pyrite, bornite, chalcocite, sphalerite, galena, and traces of cobalt, nickel, molybdenum, and silver.

### EQUILIBRIA RELATIONS DURING THERMAL METAMORPHISM OF CARBONATE ROCKS

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By the use of thermo-chemical data and by approximating certain unknown heat capacity and entropy values, the equilibrium relations ( $\Delta F = 0$ ), under ideal conditions and ignoring pressures, have been calculated for fourteen different reactions occurring during progressive thermal metamorphism of siliceous carbonates. The reactions considered involved the following minerals: calcite, dolomite, magnesite,  $\alpha$  and  $\beta$  quartz, forsterite, enstatite, diopside, wollastonite, periclase, monticellite,  $\alpha$  and  $\beta$  larnite, lime and carbon dioxide.

The calculated equilibrium temperatures at one atmosphere  $\text{CO}_2$  pressure vary from  $29^\circ \text{C.}$  for the reaction  $\text{MgCO}_3 + \text{SiO}_2 \rightleftharpoons \text{MgSiO}_3 + \text{CO}_2$  to  $877^\circ \text{C.}$  for the reaction  $\text{CaCO}_3 \rightleftharpoons \text{CaO} + \text{CO}_2$ . These equilibrium temperatures are found to be in fairly good agreement with known experimentally determined reaction temperatures. However, the consistent deviation of the experimentally determined reaction points in the lower temperature reactions from the theoretical values show that the speed at which equilibrium is attained is already strongly retarded considerably before equilibrium is reached. Due to the large time factor involved in metamorphism, the natural occurring reactions are believed to take place much closer to the theoretical than to the experimental values.

The effect of rock pressure on the equilibria temperatures at one atmosphere  $\text{CO}_2$  pressure is calculated using the Clapeyron equation by assuming that the  $\text{CO}_2$  is squeezed out of the system. In this case rock pressures of 4,000 atmospheres will cause a lowering of the equilibrium temperatures between  $40$  and  $80^\circ \text{C.}$  depending on the change in the mole volume of the solids during the reaction.

### GRAVITY OBSERVATIONS AT SEA, PART I: THE BAHAMAS ISLANDS REGION

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Simple Bouguer and free air anomalies for 234 new gravity observations in the deep water of the Bahamas Islands region are reported. Bouguer gravity anomalies for 326 points in the shallow water areas of the Bahama Islands have been provided by oil companies and oil prospecting companies. Anomaly maps are presented combining all available data. Predominantly the anomalies can be explained by simple erosion of the deep-water portions without compensation, or alternately construction of the shallow



water-portions with regional compensation. This is superimposed on a small seaward increase of gravity. A negative free-air anomaly of about 110 milligals follows the eastern boundary of the Bahamas and the southern part of the Blake Plateau. The greatest part of this anomaly can be explained by the above process, possibly complicated by boundary effects of the isostatic compensation.

### ZIRCONS AS PROVENANCE INDICATORS

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A recent study of a Scottish granite intruded into Moine "granulites" showed that while the greater part of the granite was clearly magmatic, there was considerable granitization near the contact in some areas. The magmatic origin of the granite was shown by field evidence, and by microscopic, chemical and trace-element studies; the granitized Moine could be differentiated from the normal granite without difficulty. The accessory zircons in these rocks were studied, and it was found that zircons from the granite are idiomorphic, while those from the Moines are well rounded. Furthermore, zircons from contaminated granite are similar to those from the normal granite, but they are very elongated; and zircons from the granitized Moines are recrystallized and idiomorphic, but quite distinct from those in the granite. It is suggested that this technique may have wider uses in similar studies.

### SYNTHESES AND STABILITY OF THE MUSCOVITES

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The syntheses of randomly stacked one-layer monoclinic muscovite (1Md = illite), one-layer monoclinic muscovite (1M), and two-layer monoclinic muscovite (2M) have been accomplished. The upper stability limits of muscovite are represented by a curve passing through the points 625°C.-5,000 psi water vapor pressure, 665°-15,000 psi, and 715°-30,000 psi. Above this curve sanidine + corundum + vapor are the stable phases.

The stability ranges of the muscovite polymorphs named could not be fixed accurately because of the sluggish nature of the transformations. However, the following transformations were effected: 1Md  $\rightarrow$  1M  $\rightarrow$  2M. It is believed this sequence obtains in the progressive metamorphism of sediments. The first transformation is probably dependent on factors affecting reaction rate. The second transformation may be related to a univariant curve of equilibrium (i.e., an isograd). The three-layer trigonal muscovite (3T) was obtained in only one run with 2M muscovite and its synthesis is therefore not substantiated.

Experiments on reactions alleged to represent a biotite isograd indicate that the muscovite-chlorite-quartz schists undergo the following reaction: muscovite + 2 clinocllore + 4 quartz  $\rightleftharpoons$  phlogopite + 2 cordierite + 8 water. The growth of biotite according to this reaction in low-grade metamorphic rocks must be accompanied by another phase (i.e., cordierite). Growth of biotite as a result of other reactions will not be of equivalent grade. The upper stability curve of muscovite marks the conditions of the second sillimanite (or orthoclase) isograd.

A comparison of the upper stability curve of muscovite with the minimum melting curve of the "granite" system suggests that muscovite may form in granitic magmas above approximately 25,000 psi water vapor pressure and in the solid state below that

pressure in granitic rocks. These relations may account for the apparent two generations of muscovite in some granites.

Data have also been obtained on the muscovite-leucophyllite, muscovite-paragonite, and muscovite-quartz joins.

#### THERMOLUMINESCENCE OF ARTIFICIALLY PRECIPITATED CALCITE

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A number of experiments have been performed in which calcite has been precipitated from solutions containing varying concentrations of impurity ions. Some of the impurities such as manganese and strontium appear to serve as activators for thermoluminescence. Iron generally inhibits the thermoluminescence of calcite. In some instances, specific electron trapping centers appear to be assignable to specific impurities.

A second series of experiments has been carried out in which limestones containing their natural impurities have been dissolved in acid and reprecipitated under conditions of increasing pH. These experiments indicate that certain impurities are included in the crystal lattice of calcite precipitated under specific pH conditions, since the thermoluminescence glow-curves change markedly with changing pH. It is hoped that detailed examination of these changes will yield information about the pH of the solutions from which natural calcites have been precipitated.

In order to attempt to simulate geological conditions to a small degree, some of the precipitates have been subjected to high pressures and attempts have been made to recrystallize some of the samples. Pressure effects are complex, but there appears to be a general reduction in low temperature thermoluminescence of precipitates which have been subjected to high pressure.

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## MINERALOGICAL SOCIETY (LONDON)

A meeting of the Society was held on Thursday, November 5th, 1953, at 5 p.m., in the apartments of the Geological Society of London, Burlington House, Piccadilly, W. 1 (by kind permission).

The following papers were read:—

### (1) A NEW REVIEW OF THE CHLORITES

By Dr. M. H. Hey.

The nomenclature of the chlorites and the composition boundaries between the several species and varieties are reviewed in the light of recent work. Equations are derived correlating the physical properties with the composition.

### (2) THE IDENTIFICATION AND DETERMINATION OF PLAGIOCLASE FELDSPARS BY X-RAY POWDER METHODS

By Dr. J. Goodyear and Mr. W. J. Duffin.

X-ray powder patterns have been obtained for a number of synthetic plagioclases and chemically analyzed materials of composition 0% to 100% anorthite.

The use of the x-ray powder data for the purpose of plagioclase identification and analysis is discussed.

### (3) IRON-TITANIUM OXIDE MINERALS IN ROCKS OF THE LAYERED SERIES OF THE SKAERGAARD INTRUSION, EAST GREENLAND

By Dr. E. A. Vincent and Mr. R. Phillips.

The opaque iron-titanium oxides occurring as primary phases in eight rocks of the Layered Series of gabbros and ferrogabbros have been examined with the ore-microscope, separated and chemically analyzed. The rocks all carry ilmenite in discrete crystals, associated in varying proportion with magnetites containing exsolution lamellae of ilmenite. Analyses of these magnetites show that several contain more FeO than is required to build the molecules  $\text{Fe}_3\text{O}_4$  and  $\text{FeTiO}_3$ . In those with the greatest excess of FeO, x-ray powder photographs show the lines of the magnetite pattern to be shadowed or doubled by diffuse lines due to titanospinel,  $\text{Fe}_2\text{TiO}_4$ , while exsolution bodies in this phase in the magnetite are visible with the ore-microscope at high magnifications. In other instances this exsolution has not been discerned, and it is suggested that a limited quantity of the  $\text{Fe}_2\text{TiO}_4$  molecule may remain in solid solution in the magnetite at ordinary temperatures and after slow cooling.

The contents of minor constituents in both magnetites and ilmenites—Mg, Mn, Al, Cr, V, Zn, Cu—vary fairly smoothly through the Layered Series and reflect both the ease of entry of these elements into the magnetite and ilmenite crystal lattices and their availability in the magma at each stage.

$\text{Fe}^{2+}$ ,  $\text{Fe}^{3+}$  and Ti in the magnetites vary much less regularly, and in particular the magnetites of bands very rich in opaque oxides are much richer in the  $\text{Fe}_2\text{TiO}_4$  molecule than those of adjacent average rocks from the same horizon. The composition of the final assemblage of iron-titanium oxide minerals seems to be largely determined by the equilibrium between ferrous and ferric oxides and oxygen at the time of their formation.

## (4) AN IRON-RICH FACIES OF THE BEAVER BAY DIABASE, MINNESOTA

By Dr. I. D. Muir.

This rock contains three pyroxenes, and a fayalitic olivine. From the exsolution lamellae in the pyroxenes and the inversion of part of the pigeonite to orthopyroxene, it is concluded that cooling was more rapid than for comparable rocks from the Skaergaard intrusion. The nature of the 2-pyroxene boundary is discussed.

## (5) THE REV. WILLIAM GREGOR, DISCOVERER OF TITANIUM

By Sir Arthur Russell, Bart.

The following paper was taken as read:—

(1) SPECTROGRAPHIC AND X-RAY DATA ON SOME FLUORITES  
FROM THE TRANSVAAL, SOUTH AFRICA

By Dr. J. G. D. Steyn.

Cube-edge determinations, spectrographic analyses and chemical analyses are given. It is shown that the replacement of  $\text{Ca}^{++}$  by  $\text{Sr}^{++}$  in the fluorite structure causes an increase in the cube-edge and also that some minor elements of fluorite are due to contaminating minerals present along fine cracks or as inclusions in the fluorite. The cube-edge for the purest fluorite is identical to that for synthetic fluorite of optical grade.

*(Titles and abstracts kindly submitted by G. F. Claringbull,  
General Secretary.)*

## NOTES AND NEWS

### OCCURRENCE OF "CHANNELS" IN THIN SECTIONS

BRONSON STRINGHAM AND EDWIN ROEDDER, *Department of Mineralogy, University of Utah, Salt Lake City, Utah.*

In a recent paper (this Journal, vol. 38, pp. 815-26, 1953) A. F. Frederickson states that lower index "channels" appear to crisscross some thin sections. This can be verified easily even with hand lens examination.\* The writers disagree, however, with Frederickson's interpretation of these "channels" as being features of the rock itself; we have observed them many times in the past and feel that they are merely the result of diffusion phenomena in the mounting media used to prepare the sections.

Figure 1 is a photomicrograph of one of a number of recently prepared thin sections† showing "channels," photographed using highly inclined illumination as recommended by Frederickson. The "channels" can be seen to adjoin fractures, grain boundaries, inclusions within grains, and alteration spots, and occur around the entire edge of the section. The letter "F" indicates a "channel" following a scratch accidentally put on the top surface of the slide by a grain of coarse abrasive during the final grinding; this scratch could be traced completely across the section and out onto the cooked balsam used to mount the chip. On close examination this scratch was seen to consist of a series of fractures in "chatter mark" pattern, penetrating through to the lower surface of the section. All of these "channels" appear lower in index than the adjacent material. When these slides were heated gradually and examined during the heating, the "channels" were seen to spread wider, get more diffuse, and finally disappear. They did not reappear on cooling, but when the cover glass was removed and the top of the section washed with xylol, the "channels" reappeared in the same locations and spread as much as 0.1 mm. laterally from each crack, etc., in 60 seconds, and disappeared again on heating. When the slide was warm they could be made to disappear by moving the rock section relative to the glass slide; moving the cover glass relative to the rock section had no effect. When the thin section

\* Focus on the slide using transmitted light from a high contrast area such as the edge of a window.

† These slides were prepared in the usual way, being mounted with "cooked" balsam cementing the chip to the glass slide and soft (xylol-plasticized) balsam cementing the cover. Gentle warmth was used to soften the plasticized balsam enough to squeeze out the excess without appreciably softening the cooked balsam cement.



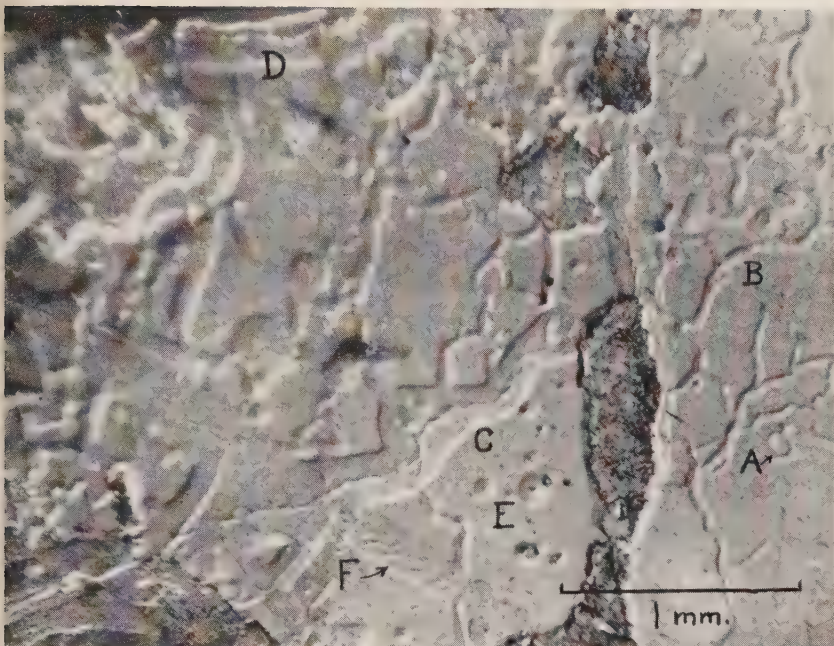


FIG. 1. Granite, Climax, Colorado. Highly inclined plane-polarized light. Light gray—quartz (polycrystalline), darker gray—partially altered orthoclase. *A*—"Channel" completely surrounding grain of quartz of different orientation than the matrix; *B*—"Channels" along quartz grain boundaries; *C*—"Channel" along fracture through quartz and feldspar; *D*—"Channel" at edge of thin section; *E*—"Channels" at holes and inclusions in quartz; *F*—"Channel" following series of "chatter marks" made across entire slide during final grinding by a stray grain of coarse abrasive.

containing the "channels" was removed from the balsam mount, washed thoroughly with xylol, dried and remounted in media having uniform index of refraction† no "channels" were visible. These data prove that the "channels" illustrated in Fig. 1 are due to xylol from the soft balsam above the section diffusing down through cracks, grain boundaries and holes, and spreading out in the cooked balsam beneath, lowering the index of this material and hence giving a low-index "channel." Not all balsam slides show them since many are heated sufficiently during the cover mounting to level out the composition gradients causing the "channels."

† The following media were tried: Methyl acrylate monomer polymerized with benzoyl peroxide; "Castolite" (methacrylate polymer); cellulose nitrate in acetone; Canada balsam in xylol; immersion liquids. When the acrylic resin mounts were washed with ethylene dichloride and when the cellulose nitrate mount was washed with acetone, "channels" formed as with balsam and xylol above.

Frederickson's statements of evidence against the phenomena of "channels" being due to mounting media (pp. 825-6) are as follows:

1. The relief features of calcite and other mineral grains change during rotation of the stage.
2. Optically polished surfaces on the thin sections did not eliminate the "channels."
3. Old slides also show "channels."
4. Several different mounting media yield the "channels."

It would appear to the authors that the variation in relief of calcite with rotation of the stage (p. 825) merely indicates that the lighting used enhances the relief features to be expected from a highly birefringent mineral like calcite and proves nothing about origin of the "channels." The presence of "channels" even with optically polished surfaces on the thin section does not eliminate the possibility of mounting media variations which could still appear as "truly an optical phenomenon." The presence of "channels" in old slides is merely a result of the use of the same mounting medium (balsam in xylol), in that diffusion boundaries in balsam, once arrested by cooling, may exist for years. As to the use of other mounting media, any media involving two materials of different index such as a resin (balsam) and solvent or plasticizer (xylol) will give gradients in index wherever composition gradients exist, and hence will show "channels."

Appreciation is expressed to the University of Utah Research Committee for their financial support of this work.

#### CHANNELS IN THIN SECTIONS—A REPLY\*

A. F. FREDERICKSON, *Washington University, St. Louis, Missouri.*

Messrs. Stringham and Roedder (1954) have commented on my paper (Frederickson, 1953a) which illustrated the fact that inclined transmitted illumination revealed much more detail in thin sections than vertical illumination techniques. It is gratifying to find that they confirm this view.

Their comments are primarily directed, however, at some of the details shown in the photomicrographs in spite of the fact that in the original paper (p. 826) it was explicitly stated that

"No attempt would be made . . . to 'explain' the origin and significance of the 'channels.'"

The word channel was enclosed in quotes and was intended to mean a passageway in the rock or mineral through which matter may have

\* Contribution No. 168 of the Department of Geology, Washington University, St. Louis, Missouri.



FIGURE 1

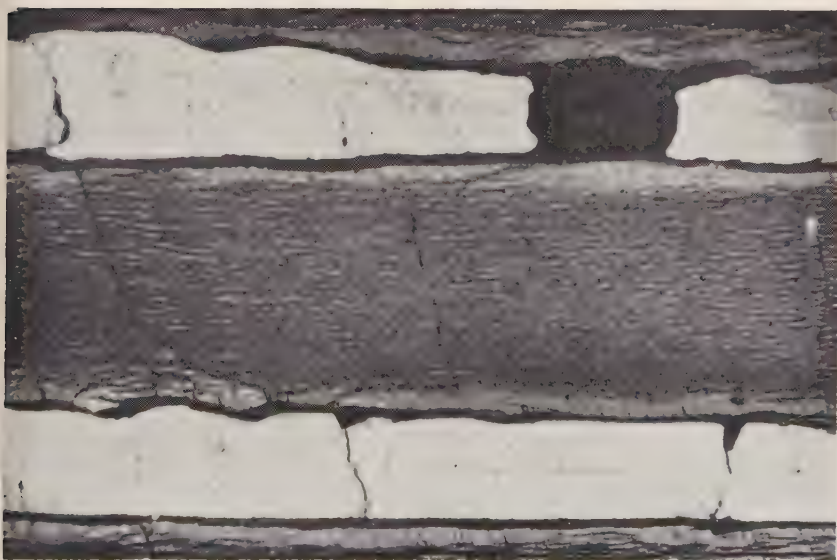


FIGURE 2



moved. They are quite correct in stating that any differences in index of refraction, including those produced by variations in the mounting media, may produce Becke lines and features much like those shown in the figures in my paper. They are not correct, however, when they deny that actual channels exist in the rocks. The alteration in Figs. 5 and 6 (Frederickson, 1953*a*) is intimately related to the crack, suture or grain boundary in the quartz (not produced during sample preparation) outlined by the channel at the top; the elliptical-shaped low-relief area in Figs. 7 and 8 outline sericite-filled holes which admitted late solutions. These are actual channels that existed long before the thin sections were prepared.

The photomicrographs included here clearly support the conclusion that much has happened to the minerals along these passageways or channels outlined by the Becke lines. Figures 1 and 2 (Magnification 73X) show quartz stringers forming a graphic texture with perthitic microcline. A double rim of recrystallized feldspar can be seen around the edges of the quartz. The rim adjacent to the quartz has a different optical orientation from the rest of the feldspar (note the black rims in the crossed-nicol photomicrograph). The next rim has the same orientation as the host microcline, but is free from perthite lamellae. Note that the cleavage cracks extend through both of the feldspar rims and abut against

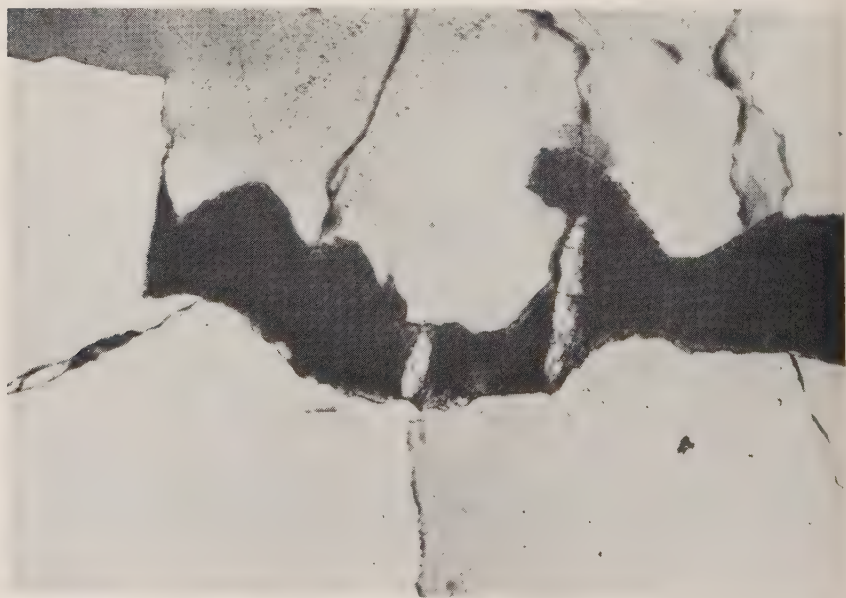


FIGURE 3

the quartz. Whether this recrystallization is due to the introduction of the quartz itself or to solutions moving along the quartz edges (along channels), is a matter which will be discussed when the petrology of these rocks is presented.

Figure 3 (Magnification 78 $\times$ ) shows a dark mineral (biotite) bounded by quartz. Numerous channels exist in the quartz. Where some of the channels cross the biotite, large, cigar-shaped clinozoisite crystals have developed. The edges of the biotite crystal are streaked. The streaking is most intense around the channels and grades inward. The color of the biotite changes from deep brown to almost colorless. Iron and magnesium have been removed from the biotite and clinozoisite crystals have developed. The spatial relationship of these secondary features to the cracks or channels outlined by Becke lines is highly suggestive that these are the passageways along which materials added to or subtracted from the minerals have moved.

Figure 4 (Magnification 157 $\times$ ) shows a rectangular zoisite crystal developing in oligoclase. The zoisite crystal is zoned. The zoisite has developed in the oligoclase at the "mouth" of a channel in the quartz. Many "streams" of what appears to be the same kind of material occurs in partings perpendicular to the twin lamellae. Some of this is zoisite but some is also micaceous, probably sericite. Tiny specks of this zoisite



FIGURE 4



and sericite-like material occurs within the channel. The passageway or channel along which some of this material has apparently moved is clearly not a fortuitous crack with a significance related only to the method of preparation of the thin section.

Diluted balsam working up along cracks or natural crystal boundaries could, and does, produce index differences that outline cracks and holes in the section. In my original paper, however, I stated that I *thought* that Becke lines outlining some channels were actually due to compositional differences in the mineral itself. I still think this is correct. However, the statement should not have been made without the accompanying evidence to support it. Some of the supporting evidence (Frederickson, 1953 *b* and *c*) occurs in other papers which have been in the hands of the editor of this Journal since September and October, 1953. The best evidence supporting this conclusion results from etching tests, using pure water under elevated pressures and temperatures, on quartz and various other minerals. Actual channels can be produced in quartz crystals (Fig. 6, Frederickson, 1953*b*) by suitably etching them with water. These channels represent zones of greater solubility than the bulk of the crystal. These channels are real: a small wire can be stuck into them. The crystals are unmounted, hence the existence of the channels cannot be questioned in terms of the method of mounting or mounting media variations. Similar channels have also been produced in albite, anorthite and microcline by hydrothermal etching techniques.

*Summary:* (1) As shown in the accompanying photomicrographs, channels or passageways along which materials or solutions have moved and which are features of the rock itself, exist as originally stated.

(2) These channels may be outlined by mounting media which can work up along them to produce some of the features shown in the photomicrographs. Some channels, however, but by no means all, are independent of the variations in the mounting media and are related to differences within the minerals themselves. Evidence documenting this opinion for quartz is now in press and details for anorthite and microcline are in preparation.

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## THE STRUCTURAL FORMULA OF AN ANTIGORITE FROM VENEZUELA

G. W. BRINDLEY, *The Pennsylvania State University, State College, Pa.*

Hess, Smith and Dengo (1952) have described a monomineralic antigorite rock from the vicinity of Caracas, Venezuela, and from a detailed chemical analysis have concluded that the composition "shows a poor fit" with the ideal serpentine formula,  $\text{Mg}_6\text{Si}_4\text{O}_{10}(\text{OH})_8$ , and is represented better by the more complex formula  $\text{Mg}_7\text{Si}_5\text{O}_{13}(\text{OH})_8 \cdot n\text{H}_2\text{O}$ , with  $n=1.4$  for the analyzed specimen, but possibly varying between 1 and 3 for other specimens of antigorite. The difficulty of reconciling this formula with the generally accepted kaolin-type structure of antigorite led the writer to re-analyze the chemical data given by Hess et al. In the writer's opinion the results obtained fit reasonably well with the ideal formula, and for the small discrepancies which are found an explanation is suggested.

It must be admitted that no unique and infallible method exists for translating a chemical analysis into a structural formula, especially in the case of silicates. Since the volume of a silicate is largely filled by O and (OH) ions, it is probably best in the first place to scale O+OH to the number of ions in the unit cell or in the structural unit which for serpentine minerals is ideally  $\text{Mg}_3\text{Si}_2\text{O}_5(\text{OH})_4$ . Table 1 shows that when the number of O atoms is scaled to 9, there are 2.038 Si ions which just exceeds the maximum permissible number by about 2%. If the chemical analysis is accepted as being strictly accurate, then the implication is that about 2% of the O and (OH) sites are vacant, which is not difficult to envisage. The chemical composition is therefore scaled to give 2 Si

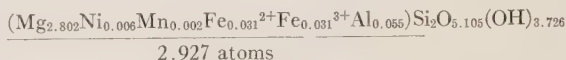
TABLE 1. COMPOSITION OF ANTIGORITE FROM NEAR CARACAS, VENEZUELA

1		2		3	
$\text{SiO}_2$	43.60	Si	2.038	2.000	
$\text{Al}_2\text{O}_3$	1.03	Al	0.057	0.055	
$\text{Fe}_2\text{O}_3$	0.90	$\text{Fe}^{3+}$	0.032	0.031	
FeO	0.81	$\text{Fe}^{2+}$	0.032	0.031	2.927
MnO	0.04	Mn	0.002	0.002	
MgO	41.00	Mg	2.855	2.802	
NiO	0.16	Ni	0.006	0.006	
$\text{H}_2\text{O}^+$	12.18	H	3.796	3.726	OH 3.726
		O	9.000	8.831	
					O 5.105

1. Composition given by Hess, Smith and Dengo, omitting traces of  $\text{Cr}_2\text{O}_3$ , CaO,  $\text{Na}_2\text{O}$ ,  $\text{K}_2\text{O}$  and  $\text{TiO}_2$ , amounting in all to 0.12.
2. Relative numbers of ions based on O=9.000.
3. Relative numbers of ions based on Si=2.000.

ions per structural unit and the results obtained are given in Table 1, column 3.

The structural formula of the antigorite can then be written



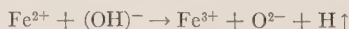
The number of octahedral ions approaches closely to 3 and the small departure from the ideal value is no greater than frequently occurs in minerals. The O ions are slightly in excess of 5 and the (OH) ions slightly below 4. The formula is in good accord with the ideal.

Various explanations may be advanced to interpret these small deviations from the ideal formula, none of which is capable of definite proof. The simplest hypothesis is that a small amount of dehydration of the mineral has occurred, which may be represented symbolically by



If sufficient water is added to bring the number of O+OH ions to 9, namely 0.169 H<sub>2</sub>O, the O:(OH) ratio becomes O<sub>4.936</sub>(OH)<sub>4.064</sub>. We cannot approach nearer to the ideal 5:4 ratio without considering why the octahedral ions depart from the ideal number 3 and the ideal change +6.

A further contributory factor to the observed composition of the mineral may be connected with the presence of ferric iron. Brindley and Youell (1953) have shown in the case of chamosite, a mineral of kaolin-type containing ferrous iron, that oxidation to a ferric form takes place without change of crystal structure according to the following reaction:



Electrical neutrality is maintained by (OH)<sup>-</sup> becoming O<sup>2-</sup>, and only H is lost from the lattice, being removed as water by aerial oxidation. This reaction also goes in the direction of increasing the O content and diminishing the (OH) content of the structure; in the present case it can be no more than a contributory factor since the amount of ferric iron is small. It should, however, be mentioned that Holzner (1938) has shown very convincingly the necessity for similar considerations in assessing the composition of iron-bearing chlorites.

In conclusion it may be stated that the composition of the antigorite discussed by Hess et al. can be reconciled satisfactorily with the simple ideal formula and that possible explanations can be given for the small deviations from the ideal composition.

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### A SIMPLE MICROSPECTROSCOPE\*

JOHN W. ADAMS, *U. S. Geological Survey, Denver, Colorado.*

The microspectroscope, or spectroscopic ocular, has been known to mineralogists for many years, but because of its limited application it has never come into common use in the mineralogical laboratory. The value of the microspectroscope in mineral identification was pointed out by Wherry (1915, 1929), who described the technique employed in its use and recorded the absorption lines and bands for a large number of minerals. The type of instrument used by Wherry consisted of a small direct-vision prism spectroscope designed to replace the conventional microscope ocular. It contained a wave-length scale, and an auxiliary prism permitted the comparison of the mineral spectra with those of

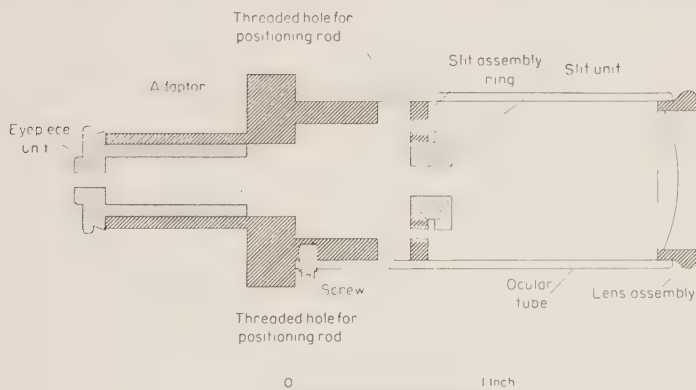


FIG. 1. Sketch showing construction of microspectroscope.

standard solutions. Microspectroscopes of that type, described and illustrated by Chamot and Mason (1948), are now difficult to obtain.

An easily constructed, inexpensive microspectroscope has been designed which the writer has found most useful in detecting the presence of rare earths in minerals. Parts from a small pocket grating spectroscope were combined with a microscope ocular as shown in Fig. 1. The upper lens of a low-power ocular from an old microscope was removed and replaced by a brass adaptor made to hold the eyepiece unit of the

\* Publication authorized by the Director, U. S. Geological Survey.

spectroscope, which contains the grating and is readily removed from the barrel of the spectroscope. The adaptor was attached to the ocular by the small screw originally used to position the ocular in the microscope tube. The unit holding the slit was unscrewed from the spectroscope barrel and fitted to a threaded ring made to slide by slight pressure inside the ocular, as shown in Fig. 1. Two holes were drilled and tapped in the ring so that small threaded steel rods could be attached temporarily and used as handles in moving the slit assembly to its correct position, which may conveniently be found by adjusting the slit assembly to obtain maximum definition of the mercury spectrum from a fluorescent lamp.

The microspectroscope can be used with either a petrographic or a binocular microscope, so that both small grains and hand specimens may be examined. Illumination, which may be by transmitted or incident white light, should be intense. A microscope lamp that can be focused is useful in directing a concentrated beam of light from above the stage. When substage illumination is desired, the upper condensing lens should be in place. It is important to remove the blue filter commonly used with microscope lamps, as the absorption spectrum of the filter may mask that of the mineral being tested.

The absence of a wave-length scale is not a serious defect, as the characteristic absorption-line pattern of many individual minerals can be recognized after a little practice using samples of those minerals described by Wherry (1929) as showing distinctive spectra.

The instrument has been found very useful in the rapid detection of monazite and xenotime in placer concentrates and in distinguishing between rare-earth minerals with similar optical properties but contrasting compositions, such as xenotime and bastnaesite.

The writer wishes to acknowledge the advice and assistance of Mr. Charles G. Bay, Chief of Property Maintenance, U. S. Geological vey, Denver, Colorado, in the construction of the instrument.

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GEIKIELITE FROM MOUNT JEMORAKLY-TUBE,  
NORTH CAUCASUS, U.S.S.R.N. EFREMOV, *New York, N.Y*

## INTRODUCTION

The investigation which led to the description of this rather rare mineral was initiated through a suggestion of Professor V. I. Kryzhanovskiy, Director of the Mineralogical Museum of the Academy of Sciences of the U.S.S.R., to whom the writer showed some grains of the mineral included in a purple chromian chlorite. A microchemical test by Prof. Kryzhanovskiy suggested that the mineral probably belonged to the ilmenite group, and might be the magnesian member, geikielite.

## OCCURRENCE

In the North Caucasus Mountains are found very extensive outcrops of serpentinite intrusions along the upper course of the Kefar-Agur River in the Great Zelenchuk Basin. The highest peak (9,000 ft.) in the region, Mount Jemorakly-Tube, is composed entirely of serpentinites. Microscopic study shows that the parent rocks of these serpentinites were lherzolites, saxonites, and, to a lesser extent, dunites.

Ore bodies of chrome-spinels, magnetite, and spinel crop out in this region. Analyses of the chrome-spinels indicate material of medium grade, containing from 45.07–54%  $\text{Cr}_2\text{O}_3$ . The ore bodies occur mainly in the immediate vicinity of Mount Jemorakly-Tube. The main chromite deposit lies on the western slope of a ridge forming the watershed between the Kefar-Agur and Kefar Rivers, at the juncture of this ridge and the north side of Mount Jemorakly-Tube. Here chromite deposits occur as uncommon lenses in a tabular, completely serpentinitized body of dunite. The main deposit, which crops out over a length of several meters, is red by a very thick mass of talus blocks.

Fractures in the chromite ores have been filled with green and purple chromian chlorites. In these the writer found several large pieces of these chlorites with rare inclusions of the then unknown mineral, now identified as geikielite. Another occurrence of geikielite in the U.S.S.R. was established by Kashin (1937), who found it in association with chlorites in the chrome-spinel deposits of the Camel Mountains, in the Southern Urals.

## DESCRIPTION

The mineral is imbedded in a matrix of chromian chlorites and forms prismatic crystals as much as 6–7 mm. in diameter. The fracture is conchoidal. The mineral is not magnetic. It is black and lustrous with a

purplish-brown streak;  $H=5$ ;  $G=4.1$ ; it is slowly soluble in HCl. In thin section it is translucent and purplish-red in color. It is uniaxial negative with  $\omega=2.35$ ,  $\epsilon=1.98$ ; absorption weak  $\omega < \epsilon$ .

The results of the chemical analysis performed in the Geochemical Laboratory of the Institute of Applied Chemistry, Rostov on Don, is given in Table 1, column 1. A comparison of the composition of this mineral with that from the Camel Mountains chromium spinelid deposits in the Southern Urals and with magnesian spinels from Ceylon is also given in Table 1.

The approximate empirical formula of the Caucasus geikielite may be written as  $(Mg, Fe, Mn)TiO_3$ . The composition may be expressed in terms of the three end-member molecules: geikielite ( $MgTiO_3$ ), crichtonite ( $FeTiO_3$ ), and pyrophanite ( $MnTiO_3$ ). The ratio of these molecules is  $MgTiO_3:FeTiO_3:MnTiO_3=31:10:1$ .

TABLE 1. ANALYSES OF GEIKIELITE

	1	2	3	4	5	6
SiO <sub>2</sub>	0.85	0.014	1.02	0.016	—	—
TiO <sub>2</sub>	57.52	0.720	58.24	0.728	63.94	61.32
Al <sub>2</sub> O <sub>3</sub>	1.22	0.012	1.04	0.010	—	—
Cr <sub>2</sub> O <sub>3</sub>	0.65	0.004	0.15	—	—	—
Fe <sub>2</sub> O <sub>3</sub>	2.82	0.017	4.05	0.025	0.25	2.03
FeO	12.30	0.171	14.96	0.208	10.09	7.75
NiO	0.27	0.003	—	—	—	—
MgO	21.57	0.539	20.36	0.501	25.79	28.95
MnO	1.27	0.017	0.24	0.003	—	—
CaO	0.32	0.05	none	—	—	—
V <sub>2</sub> O <sub>5</sub>	0.25	—	traces	—	—	—
P <sub>2</sub> O <sub>5</sub>	0.17	0.001	—	—	—	—
CO <sub>2</sub>	0.45	0.010	—	—	—	—
H <sub>2</sub> O	0.17	0.009	—	—	—	—
Total	99.83		100.06		100.07	100.05

1. Geikielite from Mount Jemorakly-Tube, North Caucasus, USSR. *Analyst* Fanny Cantor.
3. Geikielite from the Camel Mountains, Southern Urals, USSR. (Kashin, 1937); 5 and 6 geikielites from Ceylon (Doelter).

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## A CORRECTED UNIT CELL FOR BERYLLONITE

JOHN P. WEHRENBURG, *University of Illinois, Urbana, Illinois.*

The space group for beryllonite, as earlier proposed by Gossner and Besslein (1), is incorrect due to the fact that their technique (the Laue method) was limited to the recording of only the lower level reflections. By use of the Weissenberg and the Buerger precession techniques, it was found that symmetry changes occurred in the higher orders. Unit cell dimensions were taken with a 14 cm. diameter Hayes powder camera.

The correct space group is  $P2_1/n$ ; the number of molecules per unit cell = 12. The unit cell dimensions are  $a_0 = 8.16$ ,  $b_0 = 7.79$ ,  $c_0 = 14.08$ ;  $\beta = 90^\circ$ . This gives an axial ratio of  $a:b:c = 1.0475:1:1.8074$ . The  $x$ -ray density,  $\rho$ , was calculated to be 2.831.

This work was done in the  $x$ -ray laboratories of the Chemistry Department of the University of Illinois. I wish to thank Dr. George L. Clark and Mr. Walter Thatcher for the use of equipment and for their advice.

## REFERENCE

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Louis H. Ahrens, Assistant Professor of Geochemistry in the Department of Geology and Geophysics, Massachusetts Institute of Technology, has resigned to accept a Readership in Mineralogy at Oxford University commencing in January 1954. Professor Ahrens is widely known for his spectrochemical research on geological age and will continue similar work in England.

Herbert E. Hawkes, recently Chief of the Geochemical Prospecting Section of the U. S. Geological Survey, Denver Federal Center, has joined the staff of the Department of Geology and Geophysics at the Massachusetts Institute of Technology as Lecturer in Geochemistry. Dr. Hawkes will conduct one course in Geochemical Prospecting and will carry on geochemical research at the Institute.

Gordon J. F. MacDonald, Junior Fellow at Harvard University, will assume the duties of an Assistant Professor of Geology in the Department of Geology and Geophysics, Massachusetts Institute of Technology, in July 1954.

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The Geological Society of America published the following article in the October, 1953, issue of its Bulletin: "Bibliography and Index of Literature on Uranium and Thorium and Radioactive Occurrences in the United States. Part 2: California, Idaho, Montana, Oregon, Washington, and Wyoming," by Margaret Cooper of the Division of Raw Materials, U. S. Atomic Energy Commission. Since this 69 page bibliography may prove helpful to both geologists and laymen interested in uranium prospecting, the Society has prepared

reprints for public sale at 25 cents per copy. Remittance must accompany orders, which should be sent to:

*The Geological Society of America,*  
419 West 117 Street,  
New York 27, New York.

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A new bibliography series has been compiled by the Atomic Energy Commission Information Branch. The first part of this series consists of 219 selected references which appear to be of interest to industries in the metallurgical and ceramics fields. It is one of a series of bibliographies covering non-classified reports on research and development work sponsored by the A.E.C. Copies may be obtained from the Industrial Information Branch, Atomic Energy Commission, Washington 25, D. C.

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The Academy of Natural Sciences of Philadelphia has given the 1953 Hayden Memorial Geological Award, a gold medal, to Norman L. Bowen, petrologist and geophysicist of the Carnegie Institution of Washington. Dr. Bowen was cited as "a leader in the physico-chemical research in petrology, introducing systematic quantitative, experimental work on problems of origins of minerals and rocks."

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At the 66th annual meeting of The Geological Society of America and affiliated Societies held at Toronto, Canada, Nov. 9-11, 1953, the following medals and awards were presented:

Esper S. Larsen, Jr., Penrose Medalist.

J. F. Schairer, Arthur L. Day Medalist.

William F. Foshag, Roebling Medalist.

L. H. Ahrens, recipient of The Mineralogical Society of America Award.

Fritiof Fryxell, recipient of the Neil Miner Award.

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*The Editor, The American Mineralogist*

Dear Sir:

The rounding rule which appears as a footnote on page 787 of my recent paper "In defense of the second decimal" (Vol. 38, pp. 784-793, 1953) is not the one used in the numerical work leading to Tables 1 and 2, and I must apologize to readers who may have attempted to check the calculations. The correct rule is:

"5's have been rounded to the nearest *even* number."

Somewhere in the proofing and printing process the word "even" was dropped from the footnote.

FELIX CHAYES

## BOOK REVIEWS

VERFÄRBUNG UND LUMINESZENZ. BEITRÄGE ZUR MINERALPHYSIK, by KARL PRŽIBRAM. Vienna, Austria, Springer Verlag (1953). xii+275 pages, 69 figures. Price \$8.25.

After more than 30 years of fruitful research on the effect of radioactive rays on color and luminescence of minerals, Přizbram has written a welcome review of this important field of mineral physics. The field has not been reviewed since the publication in 1910 of Doelter's book, *Das Radium und die Farben*, which consisted of 133 pages dealing with early qualitative observations and contained a bibliography of 160 papers. The great difference between the two reviews, both with respect to the amount and nature of experimental data covered and to the manner of treating the data theoretically, is a measure of the advance that has been made in solid-state physics during the intervening years. Přizbram includes a bibliography of 929 papers.

The book is not a general reference for all types of mineral coloration and luminescence, but rather a specialized monograph on coloration and luminescence induced in minerals by radioactive rays in nature. As has become evident, especially from the work of Přizbram and his colleagues, many minerals show these effects of irradiation.

The book is divided into two parts. Part 1, consisting of nine chapters, deals with techniques of laboratory study of radiation effects on natural and synthetic minerals and with current theoretical interpretations of the phenomena observed. A listing of the chapter headings (freely translated) will suggest the scope of this part: historical review, methods for producing and investigating coloration, experimental data on coloration of alkali halides and other substances, photoelectric effect relative to coloration, theoretical concepts, coloration of glasses, coloration by colloids, theory of color development, and luminescence.

An ever-present difficulty in writing about a borderline field like mineral physics is the fundamental difference in outlook that divides physicists from mineralogists, as discussed by Bragg.<sup>1</sup> Přizbram is exposed to censure from both camps. In part 1 Přizbram gives the physicist's viewpoint and demonstrates the power of modern electronic theory of crystals to explain the major features of radiation coloring and luminescence. His discussion of the role of crystal imperfections—cracks, impurities, and vacant ionic sites—is especially illuminating.

In part 2 he presents a wealth of descriptive information for the mineralogist who cannot brush aside the complexities of natural minerals as tiresome details. The chapter headings of part 2 are: possibilities for coloration in nature, rock salt, other halides, fluorite, oxides and sulfides, carbonates, sulfates, nitrates and phosphates, silicates, diamond, pleochroic halos, and concluding remarks.

Among mineralogists there is increasing interest in radiation-induced fluorescence, phosphorescence, thermoluminescence, and coloration. Přizbram's good coverage and discussion of the literature on minerals will greatly help workers in the field and will encourage others to enter the field.

Of special interest to mineralogists are the sections on radioluminescence, criteria for recognizing radiation coloring, blue halite, fluorite, spodumene, and pleochroic halos. In the last section there is a reminder that the alpha-emitting nucleus that formed a halo indicative of an air equivalent range of 1.74 cm. has not yet been identified.

It is regrettable that Přizbram apparently missed seeing the account of an interesting

<sup>1</sup> Bragg, W. L., Acceptance of the Roebling Medal of the Mineralogical Society of America: *Am. Mineral.*, **34**, 238-239 (1949).



practical application of his discovery of radiophotoluminescence, namely the paper by Schulman and others<sup>2</sup> on the development of a radiation dosimeter.

The book is well bound and printed. The index is not adequate but is bolstered by the bibliography, which is also indexed. The book is a *must* for every serious mineralogical library.

K. J. MURATA,

U. S. Geological Survey, Washington 25, D. C.

CONVERSATION WITH THE EARTH, by HANS CLOOS (Translated from the German by E. B. Garside; Edited and slightly abridged by Ernst Cloos and Curt Dietz), xxx+413 pp., Alfred A. Knopf, New York, 1953. Price, \$5.75.

When *Gespräch mit der Erde* was first published in 1947 it gained enormous popularity not only as a charming autobiography and travel diary, but also as a fascinating exposition of geological method and of geological processes.

The translation preserves in large measure the interesting style, conciseness, and clarity of the original. The text has been condensed slightly, perhaps 10 percent, and the number of plates reduced from 71 to 52. The plates are smaller and not as well printed or arranged as in the German edition, but 26 explanatory maps and diagrams have been added, which aid a great deal in following descriptions of journeys and of geological processes.

Hans Cloos traveled extensively on four continents. Wherever he went he found, solved, and elucidated major geological problems. When an answer was not forthcoming from field evidence, or where more than one alternative appeared plausible, he set up and carried out intricate and ingenious experiments that usually suggested a definite solution. For example, one series of "time scale" experiments suggested that perhaps grabens like those of the Red Sea and the Rhine Valley may be structural depressions in the crowns of domes which have been produced by vertical upwelling, rather than being down-dropped trenches between two crustal blocks that have been pulled apart by horizontal forces.

These experiments explained many things for which there were previously no explanations. The work of Hans Cloos in experimental tectonics is classic, and most later work in this field is directly or indirectly based upon it.

Equally successful and impressive were the concepts developed primarily from field observations. Ideas concerning the mechanism of intrusion of granitic rocks and the development of their fabric, conceived during early work on the Erongo granite of South Africa and developed by laboratory experimentation and detailed field studies of the granite of the Riesengebirge in Silesia, were later tested on a grand scale by application to the tremendous granodiorite batholith of the Sierra Nevada. Students and followers have applied these ideas with similar success almost the world over.

There is no chapter in the book headed "Experimental Geology" or "Field Methods." It is a delightful account of a marvelously full and rewarding lifetime of field work, experimentation, and reflection woven into as pleasing a story of scientific accomplishment as can be found anywhere. It is not a systematic treatise on geology. It is not a textbook. But one who reads it carefully will come away with a better appreciation of geology, of geological processes, and of geologists than he would gain by reading many, many textbooks.

EARL INGERSON,

U. S. Geological Survey, Washington 25, D. C.

<sup>2</sup> Schulman, J. H., and others, Dosimetry of  $x$ -rays and gamma-rays by radiophotoluminescence: *Jour. Applied Physics*, **22**, 1479-1487 (1951).

HISTORICAL DEVELOPMENT OF INCLUSION THERMOMETRY, by F. G. SMITH, 149 pp. University of Toronto Press, Toronto, Ontario, 1953. Price, \$4.50.

This little volume is divided essentially into three parts:

(1) *An account of the history of the development of inclusion thermometry (Published Data)*. This section represents an extensive search of the literature for the period 1818-1952. (There is one 1953 reference.) The method of treatment is simply a chronological arrangement of brief abstracts or annotations of all of the references listed in the third part of the book. Most of the abstracts are so short that it is necessary to go back to the original articles for details of procedure and results. There is no connected account of the development of ideas, techniques, theories, or interpretations. Space could have been saved by combining parts one and three and including authors in the index.

(2) *A critical summary of the present state of knowledge*, which includes a summary of the types of inclusions in crystals, their possible modes of formation, and a discussion of the methods of studying them. The discussion is quite good, and careful attention to it will aid a novice in the field in making interpretations and will enable him to avoid many pitfalls.

At least two additional modes of formation of liquid inclusions should be mentioned: (a) Covering of etch-pits on the surfaces of crystals and (b) Covering over of solid reentrant angles between two adjacent or intergrown crystals or parts of crystals. The largest liquid inclusions known in vein minerals form in this manner.

(3) *An excellent bibliography* which will prove very useful to all workers in the field and to others interested in inclusion thermometry.

There are a few points that should be called to the attention of interested readers:

On page 52 in a discussion of Schröer's 1927 paper it is stated that "the critical temperature [of the solution(s)] was found to be somewhat above that of pure water, but the exact difference was not determined." Actually, Schröer gives values of increases in the critical temperature for solutions of alkali halides up to about 5 per cent by weight. All of the results are given to tenths or hundredths of a degree.

On page 70 the "currently accepted" figure for temperature of formation of pegmatite quartz is given as approximately 575° C. This was the accepted figure 30 or 40 years ago, but more than 10 years ago Wright himself, on whose work the figure of 575° was based, recognized that most pegmatite quartz forms below that temperature, and much of it far below.

On page 107 Smith "postulates that carbon dioxide plays a somewhat passive role." That may be true with silicates, but certainly not with quartz itself. The presence of CO<sub>2</sub> reduces the solubility of quartz markedly and causes its precipitation under conditions where it would not form if CO<sub>2</sub> were absent.

In view of the current tendency to accept the idea of important transport of material by diffusion through the lattices of minerals, it is pertinent to mention, as Smith does on page 108, that no experimental data supporting the idea have been published.

Also on page 108 he emphasizes the need for a more systematic comparison of the visual and decrepitation methods of studying liquid inclusions. One of the great needs of geologic thermometry, certainly, is a satisfactory correlation of these two methods.

On page 109 it is suggested that the pressure can be estimated by comparing the temperature obtained from thermoelectric data with temperature of decrepitation. The reviewer suggests that estimates of pressure from geologic data would be more accurate in most cases.

In the second paragraph of page 110 Smith suggests, apparently entirely on theoretical grounds, that the temperature at which glassy or "complex siliceous" inclusions become

homogeneous should approximate the temperature of formation of the enclosing crystal. This might be true of some igneous minerals that have crystallized from an essentially anhydrous magma, but even then only for certain rare cases. It will not be true of veins and other hydrothermal minerals, which are the great bulk of those studied by the inclusion method. The reviewer has heated quartz crystals containing such inclusions to temperatures that were certainly above that at which the quartz grew, yet the inclusions showed no observable change.

The method suggested on page 111, of determining temperature and pressure by varying these factors until a perfect "fit" of crystal inclusions is obtained is ingenious but would be extremely difficult, if not impossible, experimentally.

There is a short index under the heading "Bibliography (Subjects)." Many important subjects, such as "critical phenomena," "supercritical phenomena," and "critical temperature" are omitted entirely. Others are very inadequately treated. For example, there are no individual mineral names in the index. If one wants information on liquid inclusions in quartz he must look up all the references to "Pegmatitic minerals, inclusions in," of which there are 55 and all the references under "Vein minerals, inclusions in," of which there are 82.

The book was produced by the photo offset method from typed copy, but it is on good paper so that it is quite as easy to read as most printing. It is by far the most complete summary that has appeared on inclusions in minerals as clues to their temperatures of formation. It will be the starting point for researchers in this field for years to come.

EARL INGERSON,

*U. S. Geological Survey, Washington 25, D. C.*

## NEW MINERAL NAMES

### Latiumite

C. E. TILLEY AND N. F. M. HENRY, Latiumite (sulphatic potassium-calcium-aluminum silicate), a new mineral from Albano, Latium, Italy. *Mineralog. Mag.*, **30**, 39-45 (1953).

White to glassy material, massive and in elongated tabular crystals occurred in two ejected blocks of the Alban Hills, associated with hedenbergitic pyroxene, grossularite-andradite, melilite, leucite, haityne, and in one specimen also with kaliophilite. Sp. gr. = 2.93, H. = 5½-6. Analysis by J. H. Scoon (combination of 2 partial analyses) gave SiO<sub>2</sub> 28.33, Al<sub>2</sub>O<sub>3</sub> 24.67, Fe<sub>2</sub>O<sub>3</sub> 0.50, FeO 0.55, MnO 0.02, MgO 0.76, CaO 29.41, Na<sub>2</sub>O 1.11, K<sub>2</sub>O 7.20, H<sub>2</sub>O<sup>-</sup> none, H<sub>2</sub>O<sup>+</sup> 0.27, SO<sub>3</sub> 5.42, CO<sub>2</sub> 1.60, Cl 0.14; sum 99.98 - (O = Cl<sub>2</sub>) 0.03 = 99.95%. Further chemical and x-ray data are needed to determine the formula; two possibilities are X<sub>8</sub>YZ<sub>10</sub>O<sub>26</sub>(SO<sub>4</sub>) with X = 5.91 Ca, 0.40 Na, 1.73 K, Y = 0.76 Al, 0.16 Fe, 0.21 Mg, Z = 5.31 Si, 4.69 Al, SO<sub>4</sub> = 0.76, 0.40 CO<sub>3</sub>, 0.02 Cl; or X<sub>8</sub>Y<sub>4</sub>Z<sub>7</sub>O<sub>28</sub>(SO<sub>4</sub>) where X and SO<sub>4</sub> are as above, Y = 3.76 Al, 0.16 Fe, 0.21 Mg, and Z = 5.31 Si, 1.69 Al. The first would be a sheet structure, the second perhaps one of mixed groups (SiO<sub>4</sub>)(Si<sub>2</sub>O<sub>7</sub>).

Latiumite is decomposed by weak acids leaving a silica pseudomorph. It fuses before the blowpipe and the resulting glass partly devitrifies at a low red heat to a fine-grained product that gave an x-ray powder photograph similar to that of melilite.

Latiumite is monoclinic. Weissenberg and oscillation photographs gave  $a=12.12$ ,  $b=5.13$ ,  $c=10.80$  Å, beta 108°. G. calcd. 2.93. X-ray powder data are given. There is a perfect cleavage (100) and this is also the twin plane. The mineral is variable optically, indices recorded being  $\alpha=1.600$ ,  $\beta=1.606$ ,  $\gamma=1.614$ , also  $\alpha=1.603$ ,  $\beta=1.609$ ,  $\gamma=1.615$ . Mottled extinction is characteristic, with  $\alpha:c$  ranging from 16° to 28° and 2 V from 83°(+) to 72°(-). Presumably the variation in optical properties is due to variation in composition

of a solid solution. The optic axial plane is perpendicular to (010). Dispersion is marked  $r > v$ .

The name is for the locality.

MICHAEL FLEISCHER

### Cheralite

S. H. U. BOWIE AND J. E. T. HORNE, Cheralite, a new mineral of the monazite group. *Mineralog. Mag.*, **30**, 93–99 (1953).

Dark- to pale-green masses up to two inches across occur in a kaolinized pegmatite dike at Kuttakuzhi in Halkulam taluk (parish), Travancore, southern India, associated with black tourmaline, chrysoberyl, dark zircon, and smoky quartz. It also occurs sparsely in the adjacent wall-rock (kaolinized granite-gneiss) and in surface wash.

Hardness = 5, sp. gr. (pycnometer in  $\text{CCl}_4$ ) =  $5.3 \pm 0.1$  Luster resinous to vitreous, fracture uneven, brittle. Optically biaxial, positive with indices  $\alpha = 1.779$ ,  $\beta = 1.780$ ,  $\gamma = 1.816$ ,  $2V$  measured  $17.4$  to  $19.0^\circ$ . Pale green in thin section; pleochroism faint,  $\alpha$  and  $\beta$  green,  $\gamma$  green with a yellow tinge. Dispersion not perceptible. Cleavages and optical directions (tentative since crystal faces have not been identified certainly): cleavage (010) distinct, (100) difficult, parting on (001) poor,  $\alpha = b$ ,  $\gamma : c = 7^\circ$ .

A sample purified by electromagnetic separation and immersion in hot dilute HCl was analyzed by the Radiochemical Division, Chemical Research Laboratory, Teddington, giving  $\text{P}_2\text{O}_5$  26.80,  $\text{SiO}_2$  2.10,  $\text{ThO}_2$  31.50,  $\text{U}_3\text{O}_8$  4.05,  $\text{Ce}_2\text{O}_3$  14.21,  $\text{La}_2\text{O}_3$ , etc. 13.35,  $\text{Fe}_2\text{O}_3$  trace,  $\text{Al}_2\text{O}_3$  trace,  $\text{CaO}$  6.30,  $\text{PbO}$  0.92,  $\text{H}_2\text{O}^+$  0.06; sum 99.29%. The  $\text{La}_2\text{O}_3$ , etc., contained major La, Pr, Nd, minor Gd, Sm, Y. This gives  $(\text{Ca}_{1.08}\text{Ce}_{0.84}\text{La}, \text{etc.}_{0.78}\text{Th}_{1.15}\text{U}_{0.14}\text{Pb}_{0.04}(\text{P}_{3.64}\text{Si}_{0.34})\text{O}_{16})$ . A second, very similar analysis is given; this was made about 1916 by S. J. Johnstone on material supposedly from the adjacent Vilavancode taluk, but it is uncertain whether this represents another occurrence.

X-ray powder data are given for monazite ( $\text{CePO}_4$ ), huttonite ( $\text{ThSiO}_4$ ), cheralite, and synthetic  $\text{CaTh}(\text{PO}_4)_2$ ; they are isostructural and there is a systematic decrease in spacings from monazite through cheralite to  $\text{CaTh}(\text{PO}_4)_2$ . From the powder data, unit cell dimensions of cheralite are computed to be  $a = 6.74$ ,  $b = 7.00$ ,  $c = 6.43 \text{ \AA}$ , beta  $104.6^\circ$ .

The name is "from Chera, the ancient Dravidian kingdom which corresponded roughly to the modern territory of Travancore."

M. F.

### Görgeyite

HEIMO MAYRHOFER, Görgeyit, ein neues Mineral aus der Ischler Salzlagerstätte. *Neues Jahrb. Mineral.*, Monatsh. **1953**, No. 2, 35–44.

The mineral occurs with glauberite and minor halite and polyhalite in the Leopold horizon, Ischl salt deposit, Austria. Thin tabular crystals with  $c(100)$  dominant, also  $a(100)$ ,  $s(111)$ , and  $m(110)$ . Luster vitreous, colorless to yellowish,  $H. =$  to or greater than that of anhydrite ( $3\frac{1}{2}$ ).  $G. = 2.75$  monoclinic-prismatic,  $C_{2h} - 2/m$ ,  $a:b:c = 2.53:1:2.70$ , beta  $113^\circ 14'$ . Cleavage (100) distinct, fracture splintery.

The mineral is biaxial, positive,  $ns$  (Na)  $\alpha = 1.560$ ,  $\beta = 1.569$ ,  $\gamma = 1.584$ ,  $2V = 79^\circ$ . Analysis by W. Zücker gave:  $\text{SO}_3$  54.08,  $\text{Cl}$  0.06,  $\text{CaO}$  30.04,  $\text{K}_2\text{O}$  10.82,  $\text{Na}_2\text{O}$  1.77,  $\text{R}_2\text{O}_3$  0.18,  $\text{H}_2\text{O}^-$  0.34,  $\text{H}_2\text{O}^+$  2.82, gangue 0.08; sum 100.9%. This corresponds to  $\text{K}_2\text{SO}_4 \cdot 5\text{CaSO}_4 \cdot 1-1\frac{1}{2}\text{H}_2\text{O}$ , after deducting a little NaCl. Na may replace Ca or may be present as glauberite. The mineral loses little or no weight when boiled in water, but the surface becomes rough.

The name is for R. Görgey, who published many papers on the petrography of Austrian salt deposits.

DISCUSSION: The compound  $\text{K}_2\text{SO}_4 \cdot 5\text{CaSO}_4 \cdot \text{H}_2\text{O}$  has been reported as a phase in the system  $\text{K}_2\text{SO}_4\text{—CaSO}_4\text{—H}_2\text{O}$ ; see A. E. Hill, *J. Am. Chem. Soc.*, **56**, 1071–1078 (1934).  
M. F.

#### Huntite

G. T. FAUST, *Am. Mineral.*, **38**, 4–24 (1953).

#### Faheyite

M. L. LINDBERG AND K. J. MURATA, *Am. Mineral.*, **38**, 263–270 (1953).

#### Sahamalite

H. W. JAFFE, ROBERT MEYEROWITZ, AND H. T. EVANS, JR., *Am. Mineral.*, **38**, 741–754 (1953).

#### Manganpyrosomalite

CLIFFORD FRONDEL AND L. H. BAUER, *Am. Mineral.*, **38**, 755–760 (1953).

#### Hydrohausmannite

CLIFFORD FRONDEL, *Am. Mineral.*, **38**, 761–769 (1953).

#### Woodruffite

CLIFFORD FRONDEL, *Am. Mineral.*, **38**, 761–769 (1953).

#### Roentgenite

GABRIELLE DONNAY, *Am. Mineral.*, **38**, 868–870 (1953).

GABRIELLE DONNAY AND J. D. H. DONNAY, *Am. Mineral.*, **38**, 932–963 (1953).

#### Faustite

R. C. ERD, M. D. FOSTER, AND P. D. PROCTOR, *Am. Mineral.*, **38**, 964–972 (1953).

#### Moraesite

M. L. LINDBERG, W. T. PECORA AND A. L. DE M. BARBOSA, *Am. Mineral.*, **38**, 1126–1133 (1953).

#### Burbankite

W. T. PECORA AND J. H. KERR, *Am. Mineral.*, **38**, 1169–1183 (1953).

#### Calkinsite

W. T. PECORA AND J. H. KERR, *Am. Mineral.*, **38**, 1169–1183 (1953).

#### Hidalgoite

R. L. SMITH, F. S. SIMONS, AND A. C. VLISIDIS, *Am. Mineral.*, **38**, 1218–1224 (1953).

#### Montroseite

A. D. WEEKS, E. A. CISNEY, AND A. M. SHERWOOD, *Am. Mineral.*, **38**, 1235–1241 (1953).

H. T. EVANS, JR. AND STANLEY BLOCK, *Am. Mineral.*, **38**, 1242–1250 (1953).

M. F.



## DISCREDITED MINERALS

**Magnalumoxide (= Spinel)**

D. P. SERDIUCHENKO AND V. A. MOLEVA, Spinel from the Archean rocks of Southern Yakutiya. *Doklady Akad. Nauk. S.S.S.R.*, **88**, 547-550 (1953).

The material described by Bobkov and Kazitsyn (see *Am. Mineral.*, **37**, 360 (1952)) as a solid solution of  $Mg(Al, Fe)_2O_4$  was re-examined. Three new chemical analyses are given including one from the Gon River; all give  $RO:R_2O_3$  very nearly 1:1 and the optical and x-ray data correspond in every way to spinel. The analysis cited by Bobkov and Kazitsyn, which gave FeO 0.99,  $Fe_2O_3$  11.37, was apparently in error, the new analysis from Gon River giving FeO 8.23,  $Fe_2O_3$  3.28%.

N. EFREMOV

**Crestmoreite, Riversideite (= mixtures of Tobermorite and Wilkeite)**

H. F. W. TAYLOR, Crestmoreite and riversideite. *Mineralog. Mag.*, **30**, 155-165 (1953).

X-ray study and a new chemical analysis, made on type materials, show that both crestmoreite and riversideite are submicroscopic intergrowths of tobermorite ( $CaSiO_3 \cdot nH_2O$ ) and wilkeite.

MICHAEL FLEISCHER

**Belyankite (= Creedite)**

L. P. ERMILOVA AND V. A. MOLEVA, Creedite from Kazakhstan. *Doklady Akad. Nauk S.S.S.R.*, **88**, 905-908 (1953).

A new analysis, x-ray powder data and optical data, and differential thermal analysis show that belyankite is identical with creedite. Compare *Am. Mineral.*, **37**, 785-790 (1952).

M. F.

**Arequipite (= Bindheimite)**

BRIAN MASON AND C. J. VITALIANO, The mineralogy of the antimony oxides and antimonates. *Mineralog. Mag.*, **30**, 100-112 (1953).

Arequipite, described in 1878 by Raimondi as a silicoantimonate of lead, is found by optical and x-ray study of type material to be a mixture of bindheimite and quartz.

M. F.

**Coronguite (= argentine Bindheimite)**

B. MASON AND C. J. VITALIANO, *op. cit.*

Coronguite, described in 1878 by Raimondi as an antimonate of lead and silver, gave an x-ray powder photograph identical with that of bindheimite and is presumably an argentine variety, perhaps intermediate between bindheimite and stetefeldtite.

M. F.

**Flajolotite (= Tripuhyite)**

B. MASON AND C. J. VITALIANO, *op. cit.*

X-ray study shows that flajolotite (Lacroix, 1910) is identical with tripuhyite (Hussock and Prior, 1897). Since tripuhyite is found to be isostructural with bystromite, the formula  $FeSbO_4$  is more probable than  $Fe_2Sb_2O_7$ .

M. F.

**Chlorastrolite (= Pumpellyite)**

D. S. COOMBS, The pumpellyite mineral series. *Mineralog. Mag.*, **30**, 113-135 (1953).

The light bluish-green pebbles from Isle Royale, Lake Superior, named chlorastrolite by Whitney in 1847, had previously been referred to thomsonite and to prehnite, but are shown to be pumpellyite by optical, x-ray, and chemical study.

M. F.

**Buszite (= Bastnaesite)**

J. GONI AND C. GUILLEMIN, Une espèce minérale discreditée. Buszite=Bastnaésite. *Bull. soc. franc. mineral. et crist.*, **76**, 124-129 (1953).

Buszite, described in 1929 as a rare earth silicate from Khan, S. W. Africa, is shown by chemical analysis, optical and x-ray study, goniometric measurements, and differential thermal analysis to be bastnaesite.

M. F.

**Apoanalcite (= Natrolite)**

CHRISTOFFER OFTEDAHL, On "apoanalcite" and hydronephelite. *Norsk Geol. Tidsskr.*, **30**, 1-4 (1952).

This supposedly new zeolite (Ofstedahl, 1947) is shown by optical and x-ray study to consist mainly of natrolite with a small amount of another mineral, perhaps hydronephelite.

M. F.

**Zebedassite (= Saponite)**

FIORENZO MAZZI, Riesame della zebedassite. *Rend. soc. mineral. Ital.*, **8**, 134-140 (1952).

Chemical analysis, x-ray powder data, differential thermal analysis, and base exchange determinations show zebedassite to be a saponite.

M. F.

**Cryptomorphite (= Ginorite)**

M. H. HEY AND F. A. BANNISTER, The identity of cryptomorphite and ginorite. *Mineralog. Mag.*, **29**, 955-959 (1952).

X-ray, optical, and chemical data show that cryptomorphite (How, 1861) is a mixture of sodium sulfate and ginorite (D'Achiardi, 1934). The name cryptomorphite should be dropped.

**Belyankite (= Creedite)**

M. FLEISCHER, *Am. Mineral.*, **37**, 785-790 (1952).

**Cervantite, Stibianite, Volgerite,  
Hydroromeite (all = Stibiconite)**

C. J. VITALIANO AND BRIAN MASON, *Am. Mineral.*, **37**, 982-999 (1952).

**Arsenostibite (= Arsenian stibiconite)**

VITALIANO AND MASON, *loc. cit.*

**Stibioferrite (= Mixture of bindheimite and jarosite)**

VITALIANO AND MASON, *loc. cit.*

**Rivotite (= Mixture of stibiconite and malachite)**VITALIANO AND MASON, *loc. cit.***Barcenite (= Mixture of stibiconite and cinnabar)**VITALIANO AND MASON, *loc. cit.***Guadarramite (= Mixture of ilmenite and monazite)**G. SWITZER, *Am. Mineral.*, **37**, 1061 (1952).

M. F.

**Ferroschallerite (= ferroan friedelite)**CLIFFORD FRONDEL AND L. H. BAUER, *Am. Mineral.*, **38**, 755-760 (1953).**Christensenite (= tridymite)**BRIAN MASON, *Am. Mineral.*, **38**, 866-867 (1953).**Duplexite (= bavenite)**MICHAEL FLEISCHER AND GEORGE SWITZER, *Am. Mineral.*, **38**, 988-993 (1953).**Nicolayite, Hydrothorite, Maitlandite, Mackintoshite, Hyblite  
(all = thorogummite)**CLIFFORD FRONDEL, *Am. Mineral.*, **38**, 1007-1018 (1953).**Tinzenite (= manganoan axinite)**C. MILTON, F. A. HILDEBRAND, AND A. M. SHERWOOD, *Am. Mineral.*, **38**, 1148-1158 (1953).

M.F.

## NEW DATA

**Tobermorite**G. F. CLARINGBULL AND M. H. HEY, A re-examination of tobermorite. *Mineralog. Mag.*, **29**, 96-962 (1952).

A new analysis of tobermorite (Heddle, 1880) by D. I. Bothwell gave SiO<sub>2</sub> 46.17, Al<sub>2</sub>O<sub>3</sub> 4.26, FeO 0.15, MgO trace, CaO 35.15, Na<sub>2</sub>O 0.56, K<sub>2</sub>O 0.25, H<sub>2</sub>O 13.47; sum 100.01%. G. = 2.458, mean *n* 1.558. X-ray powder data show the mineral to be distinct from gyrolite and to be close to synthetic hydrated calcium silicate. The composition is regarded as CaSiO<sub>3</sub>·H<sub>2</sub>O with replacement of 3Ca by 2Al and of Ca by (Na, K)<sub>2</sub>.

M. F.

**Partzite**BRIAN MASON AND C. J. VITALIANO. The mineralogy of the antimony oxides and antimonates. *Mineralog. Mag.*, **30**, 100-112 (1953).

X-ray study of type material (Arents, 1867) showed it to be mainly composed of a bindheimite-like mineral, with quartz and limonite present. Considerable variations in index of refraction (1.61 to 1.82) and specific gravity (2.98 to 3.96) were observed; spectro-



graphic determinations showed variations, but Cu and Sb were dominant in all specimens with some Fe and Ag present, and Ca ranging from a trace to several per cent. Partzite may be regarded as the copper analogue of bindheimite.

M.F.

#### Stetefeldtite

BRIAN MASON AND C. J. VITALIANO, The mineralogy of the antimony oxides and antimonates. *Mineralog. Mag.*, **30**, 100–112 (1953).

X-ray study of type material from Belmont, Nevada (Riotte, 1867) showed that it consisted of a bindhemite-like mineral with chalcocite and pyrite. Spectrographic analysis showed that silver and antimony were the major metallic elements. Recalculation of the original analysis gives per unit cell  $\text{Ag}_{8.8}\text{Sb}_{11.4}(\text{O}, \text{OH}, \text{H}_2\text{O})_{60.6}$ . Stetefeldtite is therefore the silver analogue of bindheimite.

M.F.